

General Disclaimer

One or more of the Following Statements may affect this Document

- This document has been reproduced from the best copy furnished by the organizational source. It is being released in the interest of making available as much information as possible.
- This document may contain data, which exceeds the sheet parameters. It was furnished in this condition by the organizational source and is the best copy available.
- This document may contain tone-on-tone or color graphs, charts and/or pictures, which have been reproduced in black and white.
- This document is paginated as submitted by the original source.
- Portions of this document are not fully legible due to the historical nature of some of the material. However, it is the best reproduction available from the original submission.

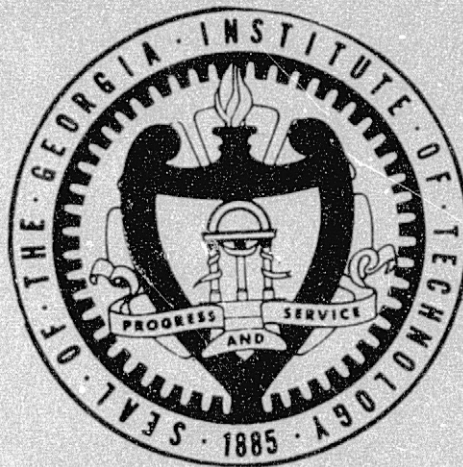
ANNUAL REPORT

NASA GRANT NSG-1288

GAS CORE REACTORS FOR ACTINIDE TRANSMUTATION

J. D. Clement, J. H. Rust, Pak Tai Wan, S. Chow

NASA Program Manager, F. Hohl



Prepared for the

National Aeronautics and Space Administration

by the

School of Nuclear Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332

April 1, 1979

(NASA-CR-158498) GAS CORE REACTORS FOR
ACTINIDE TRANSMUTATION Annual Report
(Georgia Inst. of Tech.) 75 p HC A04/MF A01
CSCI 18K

G3/73

Unclass
25110

N79-22876



ANNUAL REPORT

NASA GRANT NSG-1288

GAS CORE REACTORS FOR ACTINIDE TRANSMUTATION

J. D. Clement, J. H. Rust, Pak Tai Wan, S. Chow

NASA Program Manager, F. Hohl



Prepared for the

National Aeronautics and Space Administration

by the

School of Nuclear Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332

April 1, 1979

TABLE OF CONTENTS

	Page
LIST OF FIGURES.	iii
LIST OF TABLES	iv
SUMMARY.	1
 Chapter	
1. INTRODUCTION.	3
2. THE ACTINIDE TRANSMUTATION PROBLEM.	6
Review of Past Transmutation Studies.	9
Proposed Work	15
3. DESIGN OF REFERENCE UF ₆ ACTINIDE TRANSMUTER	19
Validity of MACH-1 Calculation for Gas Core Reactors	19
Reference Reactor Design.	21
Core Design Considerations.	21
Reflector Moderator Design Considerations	24
Actinide Blanket Design Considerations.	27
UFATR Core Neutronic Characteristics.	30
4. ANALYSIS OF ACTINIDE BURNUP IN UFATR.	33
Actinide Cross Sections	33
Computational Strategy.	35
Actinide Fuel Management During Burnup.	37
Analysis of Actinide Burnup Performance	37
Actinide Production in Core	45
CONCLUSIONS AND RECOMMENDATIONS.	48
Recommendations	48
 APPENDICES	
A. Appendix A Reprocessing Systems	
A.1 Fission Product Cleanup.	50
A.2 Actinide Reprocessing System	55
Fractionation Schemes.	58
Proposed Schemes	65

LIST OF FIGURES

<u>Figure</u>	<u>Title</u>	<u>Page</u>
2.1	Radioactivity of 1 MT of LWR UO_2 Spent Fuel	7
2.2	Toxicity of 1 MT of LWR- UO_2 Spent Fuel	7
3.1	Critical Concentration of U^{235} Gas as a Function of D_2O , and C-reflected Core Radius	20
3.2	Reactor Configuration of UFATR	22
3.3	Critical U^{233} Density and Mass vs Be Thickness	25
3.4	Neutron Leakage Fraction as a Function of Be Thickness	26
3.5	Flux Integral vs Energy Group in $\text{UF}_6 + \text{He}$ Core Region	31
4.1	Computational Strategy of UFATR	36
4.2	Plot of Avg. Flux and Power in Blanket as a Function of Burnup Time	40
4.3	Relative Blanket Fission Densities as a Function of Radial Distance at Different Times	43
4.4	Effective Multiplication of Blanket as a Function of Burnup Time	44
A.1	Fission Product Removal System	53
A.2	Actinide Reprocessing Scheme	57
A.3	Present Processing Sequence for the Removal of Actinides	63
A.4	Schematic Flowsheet of Cation Exchange Chromatographic Process for Recovery of Americium and Curium	66
A.5	Conceptual Flow Sheet for Recovery of Americium and Curium by a TALSPEAK	67

LIST OF TABLES

<u>Table</u>	<u>Title</u>	<u>Page</u>
2.1	LWR Waste Concentrations (Separate @ 10 yr. 99.5% Removal of U and Pu; per MT of Fuel)	8
2.2	Summary of Past Actinide Transmutation Studies	10
3.1	U ²³⁵ Critical Masses (kg) for a Spherical Reactor with 50 cm. Be Reflector for Different Core Radii. (T _n = 400°Q)	19
3.2	Summary of Operating Characteristics of Beginning of Life UFATR	23
3.3	Core Neutronic Parameters for Different Blanket Composition	29
4.1	Three Group Cross Section Set for the Actinides	34
4.2	Core and Blanket Parameters as a Function of Burnup	38
4.3	Gm-Atoms of the Principal Actinides as a Function of Burnup	41
4.4	Percentage of Blanket Power from Actinides as a Function of Burnup	42
4.5	Parameters RES and FAST as a Function of Burnup	42
4.6	Alpha (capture-to-fission ratio) of U ²³³ as a Function of Actinide Burnup	46
A.1	Gaseous and Fluoride Fission Products	51
A.2	Fission Product and Actinide Concentrations Leaving a LWR	59
A.3	Fission Product and Actinide Concentrations After 150 Days Storage	60
A.4	Fission Product and Actinide Concentrations Exiting from the Reprocessing Plant	61
A.5	Fission Product and Actinide Concentrations after 215 Days Storage in High Level Liquid Waste Storage Facility	62

SUMMARY

The Georgia Institute of Technology, under the sponsorship of the National Aeronautics and Space Administration, has undertaken a research program on the design and analysis of a uranium hexafluoride gas core actinide transmutation reactor (UFATR). This report summarizes results up to February 28, 1979.

One consequence of nuclear fission reactors is the accumulation of radioactive wastes. The long-term hazard of these wastes is dominated by actinides. Plutonium and uranium can be recycled within the nuclear fuel cycle, but disposal of other actinides is still a problem. If the actinides can be chemically extracted from bulk wastes, then the long-lived nuclides can be transmuted to short-lived fission products in a neutron environment. Past studies on actinide transmutation were reviewed. The UF_6 gas core reactor was selected for this application.

The core is spherical and consists of four regions. Region I is the UF_6 -He fuel mixture, region II is a beryllium reflector-moderator, region III is a liquid bismuth-actinide blanket and region IV is a graphite reflector. The gaseous fuel and liquid metal blanket are continuously circulated for heat removal, reprocessing of fission products, and refueling of depleted nuclides. For the present UFATR design, the core provides an abundant supply of thermal neutrons for transmutation use and yet is insensitive to composition changes in the blanket.

To study burnup of actinides in the blanket, a three-group cross section set was generated. The codes MACH I and ORIGEN were used iteratively to study the neutronics and depletion of the actinide blanket. An initial load of 6 metric tonnes of actinides was loaded into the blanket.

This quantity of actinides is produced by 300 LWR-years of operation. At the beginning, the core produces 2000 MWt while the blanket generates only 239 MWt. After four years of irradiation, the actinide mass is reduced to 3.9 metric tonnes. During this time, the blanket is becoming more fissile and its power rapidly approaches 1600 MWt. At the end of four years, continuous refueling of actinides is carried out and the actinide mass is held constant. Equilibrium is essentially achieved at the end of eight years. At equilibrium, the core is producing 1400 MWt and the blanket 1600 MWt. At this power level, the actinide destruction rate is equal to the production rate from 32 LWRs.

Chapter 1

INTRODUCTION

As part of its policy of supporting research and development programs which reside on the frontier of power technology, the National Aeronautics and Space Administration has sponsored work in gaseous-fueled reactors and plasma research. The original goal in research and development of the gas core reactor was to produce a space propulsion reactor capable of fast, manned expeditions to neighboring planets.⁽¹⁾ Although budgetary and policy factors terminated the development of nuclear powered propulsion engines, NASA has continued to sponsor fissioning plasma research consisting of cavity reactor criticality tests, fluid mechanics tests, investigation of uranium optical emission spectra, radiant heat transfer studies, and related theoretical work.^(2,3) Research has shown that UF_6 fueled reactors can be quite versatile with respect to power, pressure, operating temperature, and modes of power extraction.⁽⁴⁾ Possible power conversion systems include Brayton cycles, Rankine cycles, MHD generators, and thermionic diodes. Power extraction may also be possible in the form of coherent light from interactions of fission fragments with a laser gas mixture.

In addition, the International Security Office of the U. S. Energy Research and Development Administration (now the Department of Energy) has sponsored research on non-proliferating gas core reactor power plants.^(5,6) Initial studies show that fuel inventories may be a factor of 10 less than those in current U. S. power reactors.

The Georgia Institute of Technology has been engaged in various gas core reactor power plant concepts under NASA sponsorship. One concept

utilized a uranium plasma, breeder reactor employing a MHD generator for the topping cycle. (7,8) Power plant efficiencies of 70 percent are attainable with this high temperature reactor.

More recent work done at Georgia Tech involves the application of plasma and UF_6 reactors for breeding and actinide transmutation purposes. (9-11)

This report summarizes results for the design and analysis of UF_6 gas core actinide transmutation reactor.

References for Chapter 1

1. Ragsdale, R. G., "To Mars in 30 Days by Gas Core Nuclear Rocket," Astronautics and Aeronautics, 10, No. 1 (1971).
2. Thom, K., Schneider, R. T. and Schwenk, F.C., "Physics and Potentials of Fissioning Plasmas for Space Power and Propulsion," International Astronautical Federation 25th Congress, Paper No. 74087, Amsterdam (October, 1974).
3. Schwenk, F. C. and Thom, K. T., "Gaseous Fuel Nuclear Reactor Research," paper presented at the Oklahoma State University Conference on Frontiers of Power Technology (October, 1974).
4. Rodgers, R. J., Latham, T. S., and Krascella, N. L., "Investigation of Applications for High-Power, Self-Critical Fissioning Uranium Plasma Reactors," NASA CR-14508 (September, 1976).
5. Lowry, L. L., "Gas Core Reactor Power Plants Designed for Low Proliferation Potentials," LA-6900-MS (September, 1977).
6. Soran, P. D. and Hansen, G. E., "Neutronics of a Mixed Flow Gas-Core Reactor," LA-7036-MS (November, 1977).
7. Williams, J. R. and Clement, J. D., "Exploratory Study of Several Advanced Nuclear-MHD Power Plant Systems," Final Status Report, NASA Grant NGR-11-002-145, Georgia Institute of Technology, Atlanta, Georgia (March, 1973).
8. Williams, J. R. and Clement, J. D., "Satellite Nuclear Power Station - an Engineering Analysis," NASA Grant NGR-11-002-145, Georgia Institute of Technology, Atlanta, Georgia (March, 1975).
9. Clement, J. D. and Rust, J. H., "Analysis of UF_6 Breeder Reactor Power Plants," Final Report, NASA Grant NSG-1168, Georgia Institute of Technology, Atlanta, Georgia (February, 1976).
10. Clement, J. D. and Rust, J. H., "Analysis of the Gas Core Actinide Transmutation Reactor (GCATR)," Annual Report, NASA Grant NSG-1288, Georgia Institute of Technology, Atlanta, Georgia (February, 1977).
11. Clement, J. D. and Rust, J. H., "Gas Core Reactors for Actinide Transmutation and Breeder Applications," Annual Report, NASA Grant NSG-1288, Supplement No. 1, Georgia Institute of Technology, Atlanta, Georgia (April, 1978).

Chapter 2

THE ACTINIDE TRANSMUTATION PROBLEM

One consequence of large scale use of fission reactors for power production is the accumulation of radioactive wastes. The spent fuel discharged from a LWR consists of structural materials, unfissioned uranium, converted plutonium, fission products and other actinides. These actinides are formed from the neutron capture reaction of fertile and fissile isotopes. Figures 2.1 and 2.2 illustrate the radioactivity and toxicity* of spent LWR- UO_2 fuel, respectively. Up to 300 years the fission product component dominates; but from then onwards, the actinide component is dominant. Most of the actinide toxicity is due to uranium and plutonium. If the plutonium is recycled in LWRs or LMFBRs, it does not have to be considered in the waste management category. The uranium will most likely be recycled through the enrichment plant. Thus, the other actinides will be the principal contributors to the long term hazards of reactor wastes. The composition and radioactivity of the actinide portion of the high-level waste is shown in Table 2.1.

The ultimate method for the disposal of high-level radioactive wastes in the U. S. is still being evolved. For the short-lived component, it seems that ultimate storage in deep geologic formations of known characteristics (such as salt mines) remains the best method since less than one thousand years is required to reduce the activity to an innocuous level.

* The toxicity of a radioactive substance is defined as the quantity of water or air that would be required to dilute the substance to the RCG level — a level considered acceptable for ingestion or inhalation by the general public.

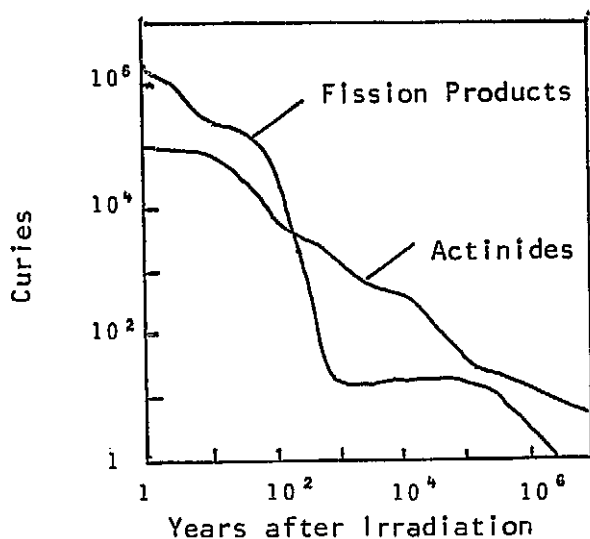


Fig. 2-1. Radioactivity of 1 MT of LWR UO_2 Spent Fuel (1)

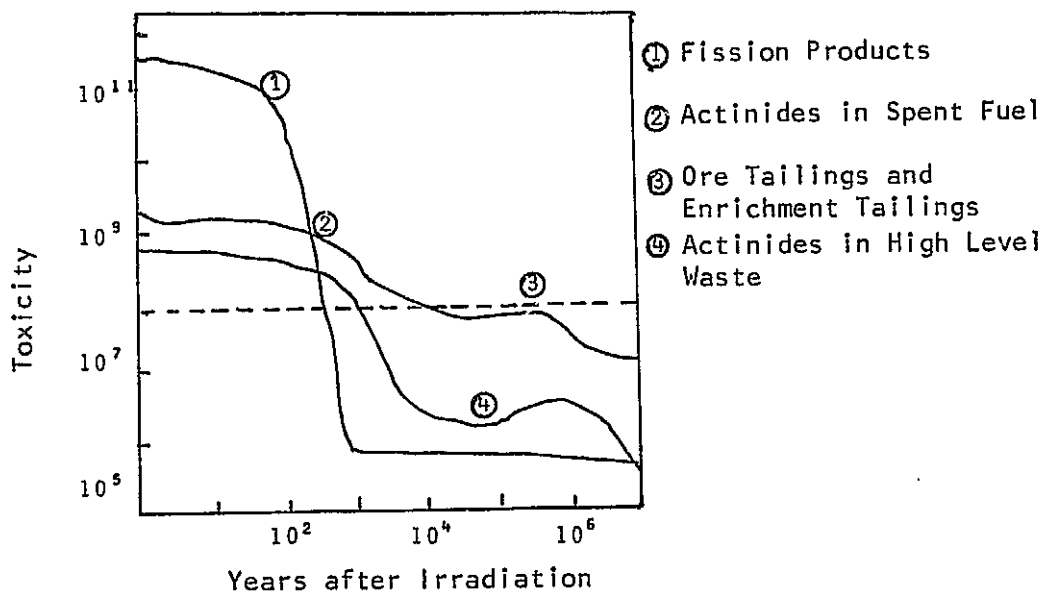


Fig. 2-2. Toxicity of 1 MT of LWR- UO_2 Spent Fuel (1)

TABLE 2.1

LWR Waste Concentrations (Separate @ 10 yr. 99.5%
Removal of U and Pu; per MT of Fuel) (1)

<u>Isotope</u>	<u>Grams</u>	<u>Curies</u>	<u>Toxicity, M³ of Water</u>
²³⁴ U	1.10	-	- - -
²³⁵ U	39.5	-	- - -
²³⁶ U	20.7	-	- - -
²³⁸ U	4730	-	- - -
²³⁷ Np	532	-	1.25 + 5
²³⁹ Np	-	13.6	1.36 + 5
²³⁴ Pu	0.709	12.0	2.39 + 6
²³⁹ Pu	23.8	1.46	2.92 + 5
²⁴⁰ Pu	10.4	2.30	4.61 + 5
²⁴¹ Pu	3.58	359	1.79 + 6
²⁴² Pu	2.07	-	- - -
²⁴¹ Am	456	1560	3.91 + 8
^{242m} Am	1.12	10.9	2.73 + 6
²⁴² Am	-	10.9	1.09 + 5
²⁴³ Am	70.9	13.6	3.41 + 6
²⁴² Cm	0.00271	3.31	6.62 + 5
²⁴³ Cm	0.0720	8.98	4.49 + 5
²⁴⁴ Cm	10.7	864	1.23 + 8
²⁴⁵ Cm	0.928	-	4.1 + 4
²⁴⁶ Cm	0.099	-	- - -
TOTAL	5910	2870	5.27 + 8

Assurance of tectonic stability for thousands of years with a very high degree of confidence is quite possible in some geologic formations. For the treatment of the long-lived component, much uncertainty exists because the effects of geologic, climatic, and other natural phenomena cannot be reliably extrapolated in the time span of thousands to millions of years. This study deals with one alternative, the neutron-induced transmutation of actinide wastes.

Review of Past Transmutation Studies

The objective of actinide transmutation is to convert the waste from an actinide waste composition to a fission product composition. The actinide elements typically have very long half lives and relatively large neutron cross sections for transmutation, especially for the fission process. After being converted to fission products, these wastes would require much shorter storage times to decay to background radiation levels.

A technical hurdle that must be overcome before actinide transmutation can become a reality is the chemical extraction of actinides at high efficiencies from bulk waste. Numerous studies have been performed on the chemical removal of actinides from high-level wastes.⁽²⁻⁵⁾ Studies to date have not been able to determine the feasibility (or infeasibility) of chemical processes for the satisfactory removal of actinides wastes. The Oak Ridge National Laboratory is currently conducting an extensive study in this area.⁽⁵⁾

Many research organizations have performed studies on transmutation using different reactor systems. A chronological list of (1) the principal investigator(s), (2) the investigator's affiliation(s), and (3) a brief description of the transmutation studies conducted is given in Table 2.2.

TABLE 2.2

Summary of Past Actinide Transmutation Studies

Investigator(s) (Organization)	Description	Reference (Date)
M. Steinberg G. Wotzak B. Manowitz (BNL)	Physics and economics of transmuting Kr-85, Sr-90, and Cs-137	6 (1964)
M. Steinberg M. V. Gregory (BNL)	Transmutation of fission product in a spallation reactor	7 (1967)
H. C. Claiborne (ORNL)	Discussion of fission product transmutation; investigation of actinide recycling in a PWR	8 (1972)
W. C. Wolkenhauer (PNL)	Physics of transmuting Sr-90 and Cs-137 in CTR	9 (1972)
W. C. Wolkenhauer B. R. Leonard, Jr. B. E. Gore (PNL)	Evaluation of potential of a CTR for transmuting fission products and actinides	10 (1973)
B. E. Gore B. R. Leonard, Jr. (PNL)	Physics of transmuting massive amounts Cs-137 in a CTR blanket	11 (1974)
K. J. Schneider A. M. Platt (PNL)	Comprehensive overview of waste management alternatives including actinide transmutation	12 (1974)
R. R. Paternoster (U. of Florida)	Calculation of actinide transmutation with a UF ₆ Gas Core reactor	13 (1974)
R. J. Breen (WARD)	Actinide transmutation rates in oxide and carbide fueled LMFBR	14 (1975)
S. Raman (ORNL)	Review of actinide transmutation in many devices	15 (1975)
S. Raman C. W. Nestor, Jr. J. W. T. Dabbs (ORNL)	Actinide transmutation in a U ²³³ -Th ²³² reactor	16 (1975)
A. G. Croff (ORNL)	Review of actinide transmutation studies	17 (1975)
A. G. Croff (ORNL)	Parametric survey of actinide transmutation	18 (1976)

TABLE 2.2

Summary of Past Actinide Transmutation Studies (cont'd)

S. L. Beaman E. A. Aitken (GE)	Physics of recycling wastes from 3 BWRs and 1 LMFBR in an LMFBR	19 (1976)
J. J. Prabulos (GE)	Calculation of actinide transmutation in a 1500 MWe carbide fueled LMFBR	20 (1976)
W. Bocola L. Frittelli G. Grossi A. Moccia L. Tondinelli (CNEN-CSN, Italy)	Calculation of sensitivities of actinide buildup to cross section changes; comparison of risks from nuclear transmutation and geologic disposal	21 (1976)
T. A. Parish E. L. Draper, Jr. (U. of Texas)	Engineering and physics design of a CTR for long-lived fission product transmutation	22 (1976)
R. H. Clarke G. A. Harte (GEGB, UK)	Actinide production and transmutation in MAGNOX and sodium cooled fast reactors	23 (1976)
R. P. Rose (EPRI)	Engineering and physics design of a tokamak fusion actinide transmuter	24 (1976)
U. P. Jenquin B. R. Leonard, Jr. (PNL)	Physics of transmuting actinides in CRT blankets	25 (1976)
D. H. Berwald (U. of Michigan)	Engineering and physics design of a laser driven fusion actinide transmuter	26 (1977)
T. H. Pigford J. Choi (U. C. Berkeley)	Calculation of approach-to-equilibrium times for PWR and LMFBR as actinide transmuter	27 (1978)
J. D. Clement J. H. Rust (Ga. Tech)	Analysis of gas core actinide transmutation reactor	28 (1977)
G. Oliva G. Palmiotti M. Salvatores L. Tondinelli (Italy)	Comparison of actinide transmutation in LWRs and LMFBRs	29 (1978)
J. D. Clement J. H. Rust (Ga. Tech)	Design of plasma core reactors for actinide transmutation	30 (1978)

The list is restricted mainly to studies with fission and fusion reactor systems. Those interested in other systems, such as accelerator or nuclear explosive transmutation, are referred to Ref. 12, which gives a discussion of these transmutation devices and an extensive list of references.

The effectiveness of an actinide transmutation system depends on numerous factors. The principal ones are (i) neutron flux level, (ii) neutron energy spectrum, and (iii) logistics of the transmutation strategy.

The most important parameter affecting actinide transmutation rates is the neutron flux in the actinide region. All studies strive to maintain as high a flux as possible. Studies using commercial power reactors as transmuters are hampered by fixed flux levels determined by power production considerations. Typical LWR thermal fluxes are on the order of 10^{13} to 10^{14} n/cm²-sec. Typical LMFBR fast fluxes are on the order of 10^{15} to 10^{16} n/cm²-sec. For fusion reactors, Rose⁽²⁴⁾ indicated that a high neutron wall loading (about 10 MW/m²) is required for effective transmutation rates. However, tokamak fusion reactors probably cannot achieve such high wall loadings due to high plasma beta stability considerations⁽²⁴⁾ and laser driven fusion reactors will be required.

Complications may also arise due to changing characteristics of the actinide region. As actinides are irradiated, they are fissioned or converted to higher actinides by capture. Hence, the composition of the actinide mix is gradually changing with time. Initially, it consists mostly of Np²³⁷, Am²⁴¹, and Am²⁴³. Upon irradiation, some are converted to nuclides with large fission cross sections. This may cause problems because the neutron flux is usually set at the maximum permissible value

consistent with thermal hydraulic constraints. As the actinide mix becomes more fissile, the neutron flux may have to be lowered to maintain a constant volumetric heat generation rate. Upon further irradiation, fission product poisons become dominant and the flux may have to be readjusted.

The energy spectrum of neutrons irradiating the actinides is a significant factor. Many authors^(8,15,19) stated that fast reactors are superior to thermal reactors because the fission-to-capture ratio is generally higher for fast reactor neutron spectra. Rose found that thermal spectrum actinide burner concepts have difficulty achieving a high k_{eff} (about 0.85 - 0.95), whereas fast burners can attain such high neutron multiplication. However, on the basis of reaction rates, a study by Oliva, et al.⁽²⁸⁾ showed that LWRs are better than LMFBRs. This is because the fast neutron fluxes of present day LMFBRs are not large enough to compensate for the drop in neutron cross sections at fast energies so that their product, i.e. the reaction rate, is less than that of the LWR case. One clear advantage that fast reactors have over thermal reactors is that their criticality is less sensitive to the introduction of foreign materials in the core. This means that for the same reactivity penalty, larger quantities of actinides can be inserted in fast reactors and that these actinides can have more fission product impurities. For fusion reactors, the mean energy of neutrons emerging from fusion reactions is very high (14 MeV for the deuterium-tritium reaction). Theoretically, a greater number of neutron reactions, e.g. (n,2n), (n,3n), (n,p) is available as transmutation channels. In practice, the cross sections of these high energy reactions are small and they were found to contribute insignificantly to the overall reaction rates.

In fact, many fusion transmutation studies utilize well-moderated actinide blankets to maximize transmutation rates.

Another major factor affecting the overall effectiveness of actinide transmutation is the logistics of the transmutation strategy. Some studies make the simplifying assumption that actinides are loaded into the transmuter once and for all, and that they are irradiated continuously for long periods of time (typically 30 years) with no reprocessing. Under such a strategy, the actinide inventory in the transmuter will decrease almost exponentially. Some studies utilize the concept of actinide recycling. The irradiated actinides are discharged to reprocessing after one cycle of irradiation. At reprocessing a fresh batch of actinides is added to the unfissioned actinides. Together, they are extracted, made into forms suitable for irradiation and inserted back into the transmuter. After many cycles, an equilibrium is reached. From then onwards, the quantity of actinides removed in one cycle is equal to the quantity of fresh actinides added during reprocessing. For actinide recycling schemes, the actinide extraction efficiency is of vital importance. Since each time the actinides pass through the reprocessing step, a fraction is lost to waste storage together with the fission products. Consequently, these actinides are not transmuted and contribute to the long-term hazard of storage wastes. On the other hand, if all actinides are kept within the transmutation system, they will eventually be beneficially transmuted. A subtle point that affects the overall transmutation rates concerns whether converted uranium and plutonium are removed during reprocessing. During reprocessing, the fission products are removed. In the studies of Claiborne⁽⁸⁾ and Beaman,⁽¹⁹⁾ the converted uranium and plutonium are also

removed. The nuclides removed are mostly Pu^{238} , formed from neutron capture of Np^{237} and decay of Am and Cm isotopes. For such a transmutation strategy, there will be two main pathways for removal of actinides. One is via direct fission during irradiation and the other is via reprocessing as converted uranium and plutonium. Claiborne's data showed that in one equilibrium cycle, about 35% of the in-core actinides are removed—12% is fissioned directly and 23% is removed in reprocessing. The extracted Pu^{238} can be used as a breeding material for Pu^{239} . From the point of view of ultimate waste disposal, the removal of Pu^{238} constitute a postponement since Pu^{238} is a highly hazardous nuclide with toxic decay daughters. A proper disposal strategy must be developed for the extracted Pu^{238} .

Proposed Work

There is a need for the study of fission reactors specifically designed to burn actinides. As actinide transmuters, commercial power reactors have two shortcomings. The flux level is limited by power production considerations and the number of reactors serviced by one power reactor is small. Consequently, many power reactors would have to be used as transmuters. Fusion reactors do produce an abundant supply of high-energy neutrons. However, a considerable amount of basic research and developmental work is required before fusion reactors can be expected to be commercially available. Hence, there is motivation to use near-term technology to design fission reactors, especially engineered for the burnout of actinide elements. The present study is concerned about the design and analysis of a uranium hexafluoride gas core reactor for such as application.

References for Chapter 2

1. Jenquin, U. P. and Leonard, B. R., Jr., "Evaluations of Fusion-Fission (Hybrid) Concepts: Transmutation of High-Level Actinide Waste in Hybrids," Part B, EPRI-ER-469 (1976).
2. Bond, W. D., Claiborne, H. C. and Leuze, R. E., "Methods for Removal of Actinides from High-Level Wastes," Nuclear Technology, 24, 362 (December, 1972).
3. Bond, W. D. and Leuze, R. E., "Feasibility Studies of the Partitioning of Commercial High-Level Wastes Generated in Spent Fuel Reprocessing: Annual Progress Report for FY-1974," ORNL-5012 (January, 1975).
4. Bond, W. D. and Leuze, R. E., "Removal of Actinides from High Level Wastes Generated in the Reprocessing of Commercial Fuels," Transplutonium Elements, 423 (1976).
5. Blomeke, J. O. and Tedder, D. W., "Actinide Partitioning and Transmutation Program Progress Report for October 1, 1976 to March 31, 1977," ORNL-TM-5888 (June, 1977).
6. Steinberg, M., Wotzak, G. and Manowitz, B., "Neutron Burning of Long-Lived Fission Products for Waste Disposal," BNL-8558 (September, 1964).
7. Gregory, M. V. and Steinberg, M., "A Nuclear Transformation System for Disposal of Long-Lived Fission-Product Waste in an Expanding Nuclear Power Economy," BNL-11915 (November, 1967).
8. Claiborne, H. C., "Neutron-Induced Transmutation of High-Level Radioactive Waste," ORNL-TM-3984 (December, 1972).
9. Wolkenhauer, W. C., "The Controlled Thermonuclear Reactor as a Fission Product Burner," BNWL-SA-4232 (1972).
10. Wolkenhauer, W. C., Gore, B. F. and Leonard, B. R., Jr., "Transmutation of High-Level Radioactive Waste with a Controlled Thermonuclear Reactor," BNWL-1772 (1973).
11. Gore, B. E. and Leonard, B. R., Jr., "Transmutation of Massive Loadings of Cesium-137 in the Blanket of a Controlled Thermonuclear Reactor," Nuclear Science and Engineering, 53, 319-323 (1974).
12. Schneider, K. J. and Platt, A. M., "High-Level Radioactive Waste Management Alternatives," BNWL-1900, Battelle Pacific Northwest Laboratories (May, 1974).

13. Paternoster, R. R., "Radioactive Waste Disposal by Nuclear Transmutation in a UF_6 Gaseous-Core," M.S. Thesis, U. of Florida (1974).
14. Breen, R. J., "Elimination of Actinides with LMFBR Recycle," Trans. Am. Nucl. Soc., 21, 262 (June, 1975).
15. Raman, S., "Some Activities in the United States Concerning the Physics Aspects of Actinide Waste Recycling," Proc. IAEA Advisory Group Mtg. Transactinium Isotope Nuclear Data, CONF-751104-4 (1975).
16. Croff, A. G., "Actinide Transmutation Studies: A Review," Trans. Am. Nucl. Soc., 23, 545 (June, 1976).
17. Croff, A. G., "Parametric Studies Concerning Actinide Transmutation in Power Reactors," Trans. Am. Nucl. Soc., 22, 345 (1975).
18. Raman, S., Nestor, C. W., Jr. and Dabbs, J.W.T., "The U^{233} - Th^{232} Reactor as a Burner for Actinide Wastes," CONF-750303-36 (1975).
19. Beaman, S. L. and Aitken, E. A., "Feasibility Studies of Actinide Recycling in LMFBRs as a Waste Management Alternative," CONF-760622-48 (1976).
20. Prabulos, J. J., "Actinide Destruction in a 1500 MWe Carbide Fueled LMFBR," Trans. Am. Nucl. Soc., 23, 548-549 (June, 1976).
21. Bocola, W., Fritelli, L., Gera, F., Grossi G., Moccia, A. and Tondinelli, L., "Considerations on Nuclear Transmutation for Elimination of Actinides," IAEA-SM-207/86 (1976).
22. Parish, T. A. and Draper, E. L., Jr., "Determination of Procedures for Transmutation of Fission Product Wastes by Fusion Neutrons," UTNRL-FU101, U. of Texas (1976).
23. Harte, G. A. and Clarke, R. H., "Investigation into the Use of a Fast Breeder Reactor to Incinerate Actinide Waste from the U.K. Nuclear Power Programme," CEGB-RD/B/N-3903 (1976).
24. Rose, R. P., "Fusion-Driven Actinide Burner Design Study," EPRI-ER-451 (1976).
25. Berwald, D.H., "Preliminary Design and Neutronic Analysis of a Laser Fusion Driven Actinide Waste Burning Hybrid Reactor," Ph.D. Thesis, Dept. of Nuclear Engineering, U. of Michigan (1977).
26. Pigford, T. H. and Choi, J., "Actinide Transmutation in Fission Reactors," Trans. Am. Nucl. Soc., 27, 450-451 (1977).
27. Clement, J. D. and Rust, J. H., "Analysis of the Gas Core Actinide Transmutation Reactor (GCATR)," Georgia Institute of Technology, NASA Grant NSG-1288 (1977).

28. Oliva, G., Palmiotti, G., Salvatores, M. and Tondinelli, L., "Elimination of Transuranium Elements by Burnup in a Power Fast Breeder Reactor," Nuclear Technology, 37 (March, 1978).
29. Clement, J. D. and Rust, J. H., "Gas Core Reactors for Actinide Transmutation and Breeder Applications," Georgia Institute of Technology, NASA Grant NSG-1288 (1978).

Chapter 3

DESIGN OF REFERENCE UF_6 ACTINIDE TRANSMUTER

Validity of MACH-1 Calculation for Gas Core Reactors

The MACH-1 code⁽¹⁾ was chosen to perform neutronic calculations. MACH-1 is a one-dimensional, multi-group, diffusion code. It has one thermal group. At Georgia Tech, it uses a 26-group cross section set derived from the ABBN set.⁽²⁾

Due to the low density of fuel in gas core reactors, there is some doubt as to whether a simple code like MACH-1 can describe the neutronics accurately. The calculations of Mills^(3,4) were chosen as the standard. Mills used a multigroup S_n theory code. He obtained good agreement between calculation and experimental data for low fuel density reactors. Figure 3.1 shows plots of critical concentration and critical masses of U^{235} gas as a function of D_2O , Be, and C—reflected reactors as a function of core radius. Table 3.1 shows U^{235} critical masses for a spherical reactor with 50 cm of Be reflector as calculated by MACH-1.

TABLE 3.1

U^{235} Critical Masses (kg) for a Spherical Reactor with 50 cm.
Be Reflector for Different Core Radii. ($T_n = 400^\circ C$)

	Radius						
	20 cm.	30 cm.	40 cm.	50 cm.	60 cm.	100 cm	300 cm.
MACH-1 result	4.44	2.10	2.34	2.85	3.49	7.09	46.6

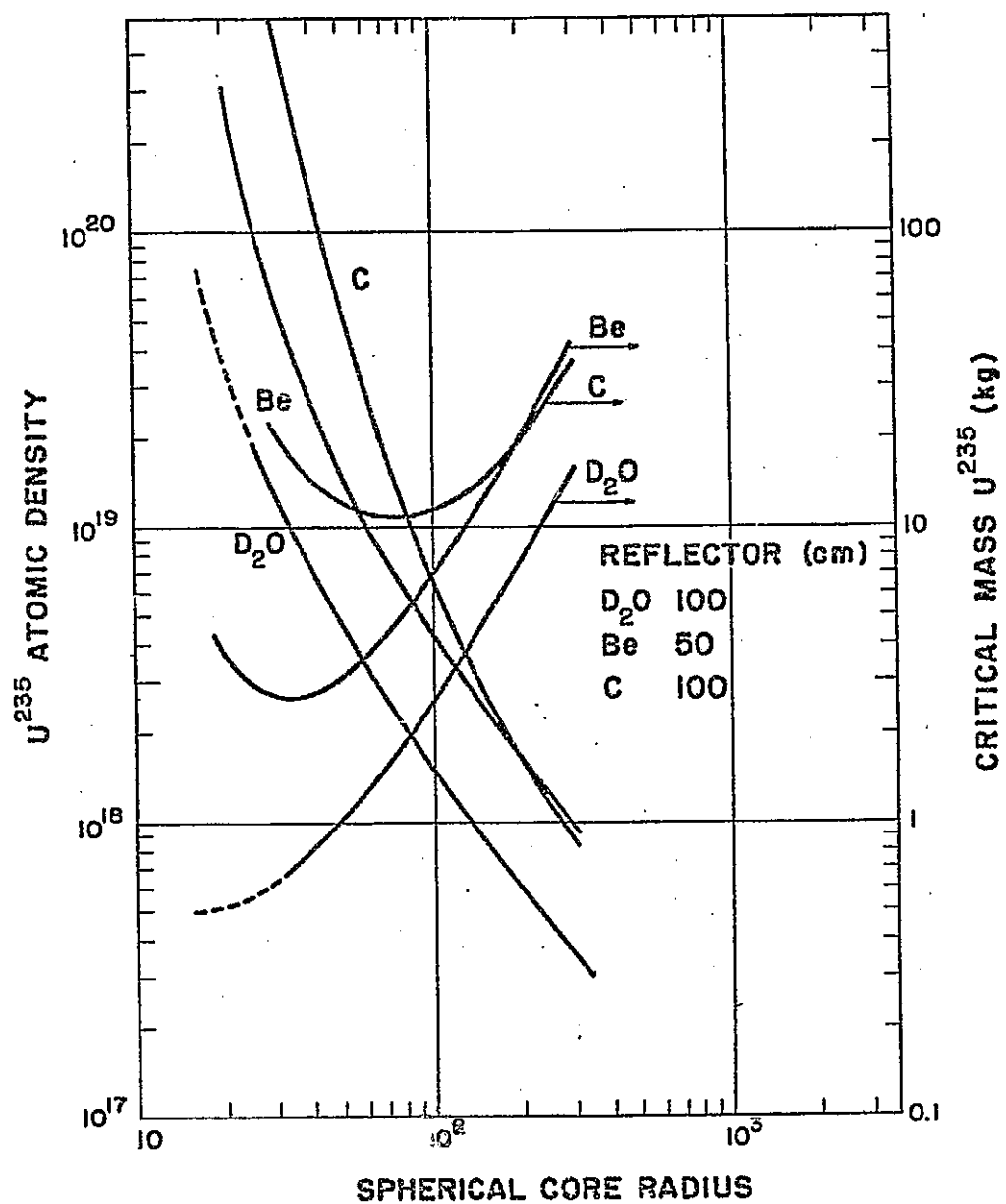


Fig. 3.1. Critical Concentration of U^{235} Gas as a Function of D_2O , Be, and C-reflected Core Radius.

For treatment of the thermal group cross sections in MACH-1, the Wescott⁽⁵⁾ formulation is followed. The MACH-1 results well match the shape of Mills curves. By adjusting the effective neutron temperature, good agreement (<10% discrepancy) is obtained.

Reference Reactor Design

The configuration of the UF_6 transmuter is shown in Fig. 3.2. It consists of four regions. Region I is the gaseous fuel region with a mixture of UF_6 and He as fuel. Region II consists of the beryllium reflector-moderator. Region III consists of the liquid bismuth-actinide blanket. Region IV is the graphite reflector. Table 3.2 is a summary of the operating reactor parameters.

Core Design Considerations

Spherical geometry is chosen for simplicity of design. A fuel mixture of UF_6 and He is used. The uranium is essentially 100% U^{233} . Since UF_6 is a very poor heat transfer agent, helium is added to improve the overall heat transfer characteristics of the mixture. Addition of helium helps to maintain a small inventory of U-233 in the heat exchangers. The fission energy is deposited in the UF_6 -He mixture in reactor core. It is pumped out of the core through heat exchangers where the fission energy is transferred. The fuel mixture is passed through reprocessing and refueling systems, where fission product poisons are removed and fresh UF_6 fuel added.

Since the ABBN cross section set does not have cross sections for helium and fluorine, these were generated from cross section data from BNL-325.^(6,7) The formalism is described previously.⁽⁸⁾ It is estimated that due to the low neutron cross sections and density of helium and fluorine, they do not effect the neutronics calculations significantly.

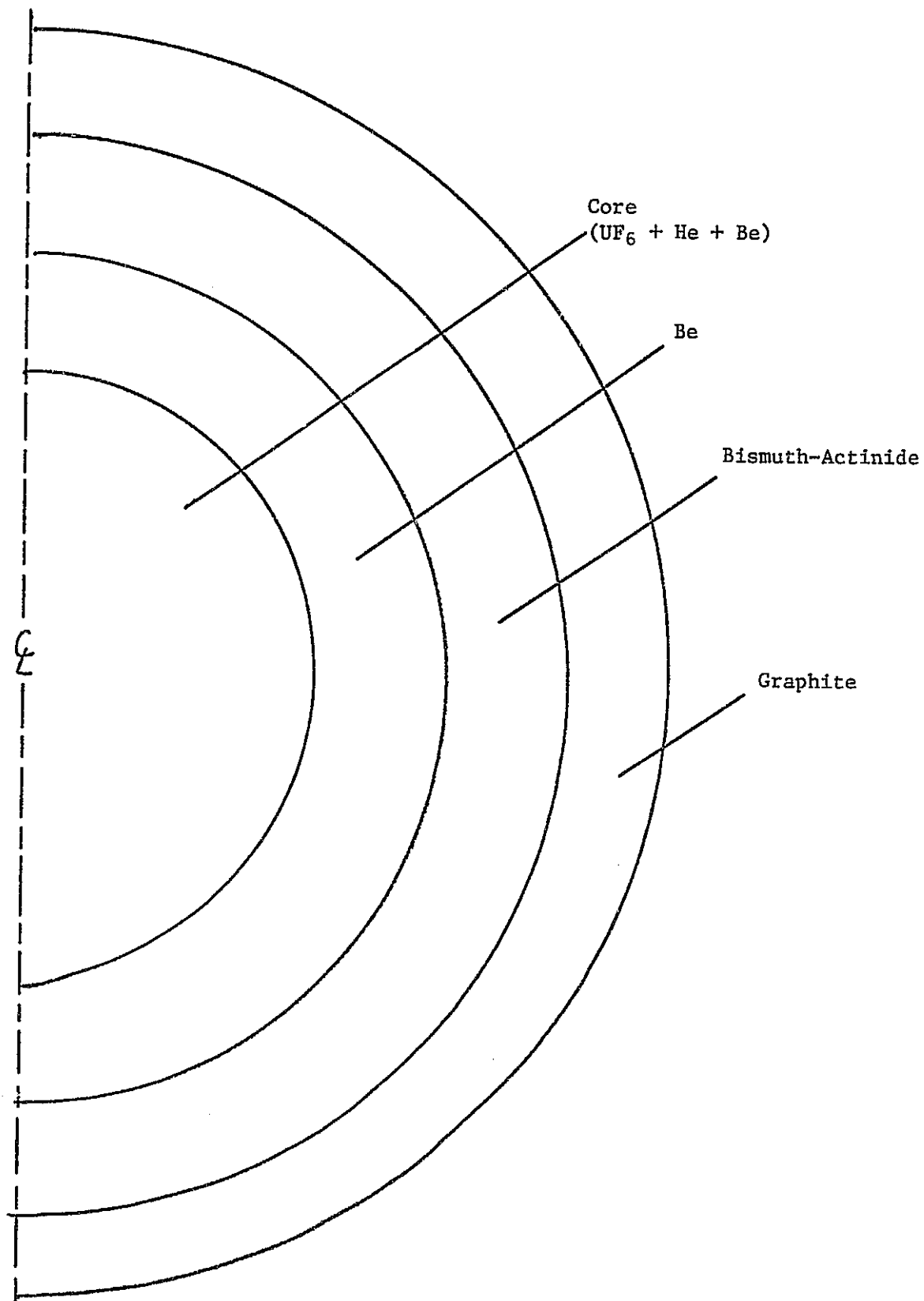


Fig. 3.2. Reactor Configuration of UFATR

TABLE 3.2

Summary of Operating Characteristics of
Beginning of Life UFATR

Reactor Type: Thermal

Geometry : Spherical

Region I:

Fuel: UF₆, He gas mixture
Enrichment: 100% U²³³
Radius: 133 cm
Core pressure: 101 atm.
UF₆ partial pressure: 1.9 atm
He partial pressure: 99.1 atm
Core power: 2000 MWt
Power density: 203 Watts/cc
Mean core temperature: 783°K(510°C, 950°F)
U²³³ critical mass: 7.05 kg
Peak to avg. power density ratio: 1.002
Avg. thermal flux: 1.16×10^{16} n/cm²-sec

Region II:

Reflector-Moderator: Be solid
Thickness: 35 cm
Mean temperature: 783°K(510°C, 950°F)
Be mass: 18.3 MT

Region III:

Blanket material: liquid Bi and actinides
Composition: 93 wt% Bi
7 wt% Actinides
Thickness: 20 cm
Mean temperature: 723°K(450°C, 842°F)
Actinide mass: 6.0 MT
Bi mass: 78.5 MT

Avg. thermal flux: 4.11×10^{13} n/cm²-sec
Power: 239 MWt
Maximum design power: 1600 MWt
Maximum design power density: 200 Watts/cc

Region IV:

Reflector material: solid graphite
Thickness: 100 cm
Mean temperature: 723°K(450°C, 842°F)
Graphite mass: 116 MT

The maximum core power is set at 2000 MWt. The core dimension is chosen such that a reasonable volumetric heat generation rate of 200 watts/cc is obtained. No detailed thermal-hydraulic calculations are performed. Knowledge of the fission density distribution in the core is required for such calculations. However, no major difficulties are anticipated in this area.

The limiting materials problem in a UF_6 core reactor is the corrosion of the core containment vessel. Since the maximum temperature of the UF_6 in the core is rather low ($> 800^\circ\text{K}$), Ni, Al, Mg, and Zr metals all have excellent F_2 corrosion resistance.⁽⁹⁾ These metals can be used as a thin liner or clad. Even if the Be is exposed to F_2 through cracks, pinholes, etc., the BeF_2 that forms when Be reacts with F_2 is reported⁽¹⁰⁾ to passivate the surface.

Reflector Moderator Design Considerations

Because Be has a high scattering cross section, a high atomic density, and the lowest absorption cross section of all metals, it is chosen as the reflector-moderator for the core.

Two conflicting considerations enter into the choice of reflector thickness. In order to have an abundant supply of core neutrons for transmutation, a thin beryllium region is desirable. However, too thin a reflector makes the core very sensitive to changes in the blanket region. Figure 3.3 is a plot of U^{233} concentration as a function of beryllium reflector thickness. The steep slope of the curve for a Be thickness less than 20 cm indicates the gas core is extremely sensitive to external moderation. For thicknesses greater than 50 cm, the core is close to an infinitely reflected assembly. Figure 3.4 shows the neutron leakage from the beryllium reflector. A Be thickness of

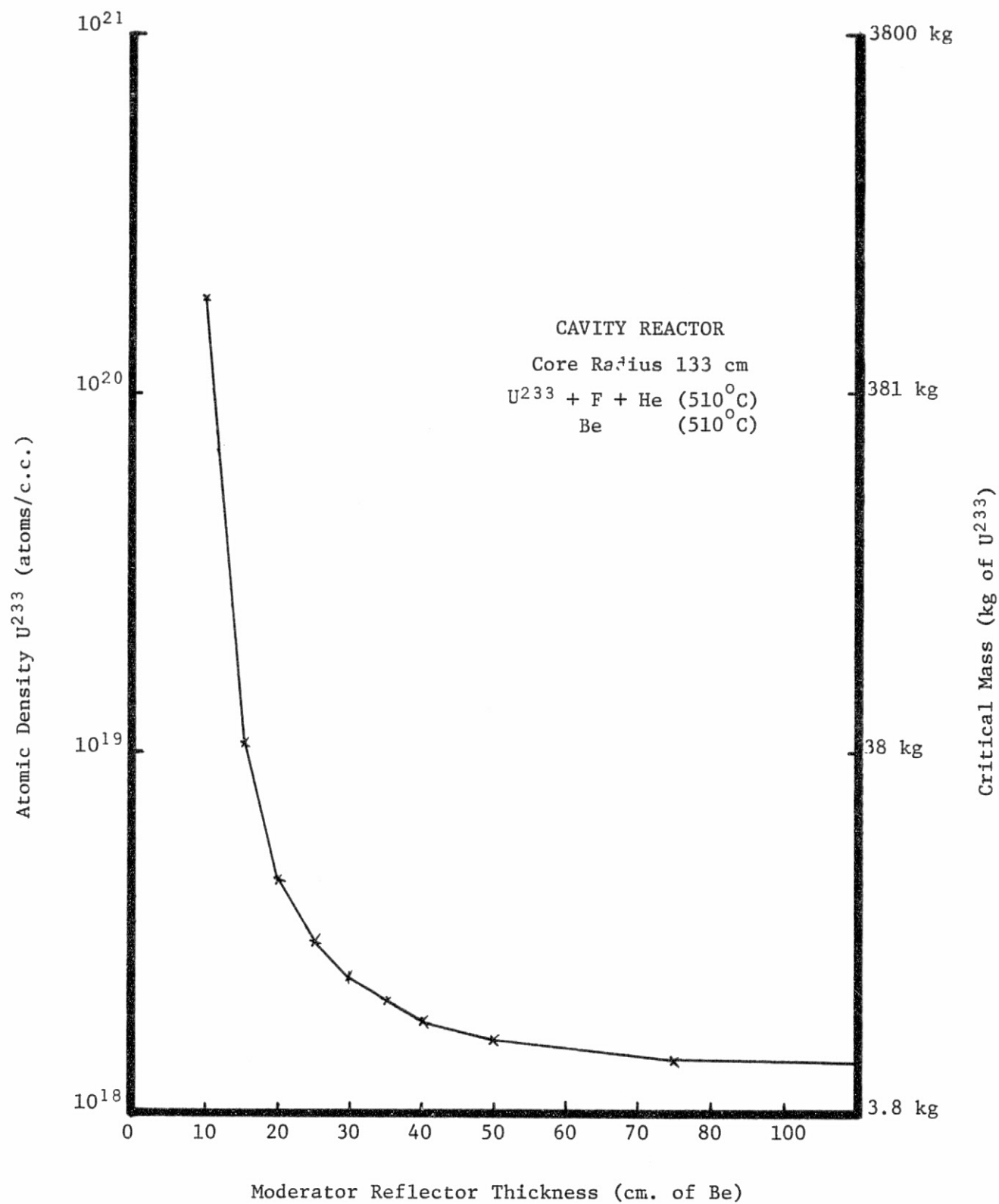


Fig. 3.3. Critical U^{233} Density and Mass vs Be Thickness

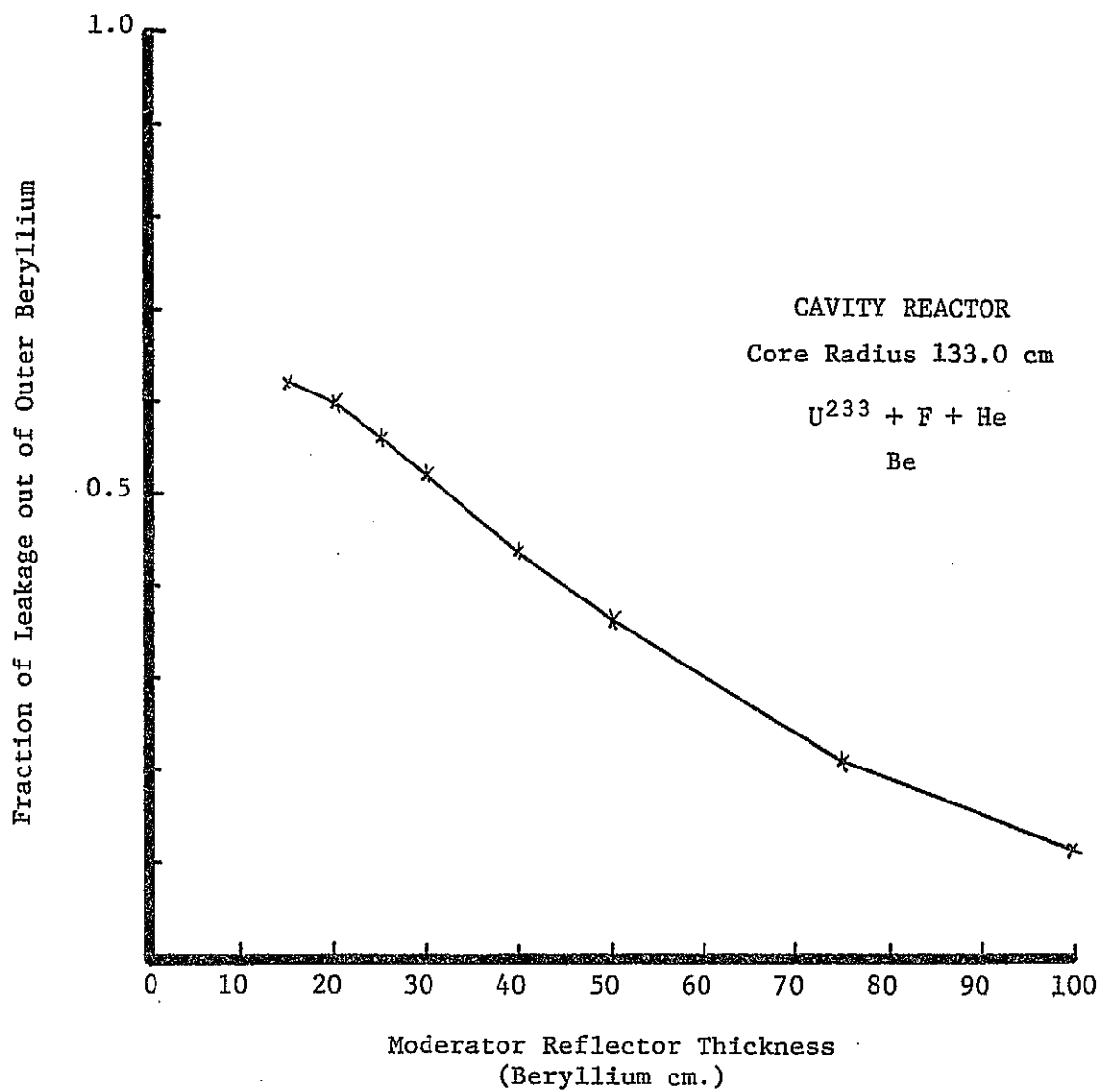


Fig. 3.4. Neutron Leakage Fraction as a Function of Be Thickness

35 cm is chosen as the optimum reflector thickness. For such dimensions, there is sufficient moderation and reflection of neutrons that the core is not sensitive to changes in the actinide blanket region. The fraction of neutrons leaked from the beryllium is 0.48, and 99.6% of these neutrons are thermalized. For a core power of 2000 MWt, 1.56×10^{20} neutrons/sec leave the core and are available for transmutation use. As pointed out by Safonov,⁽¹¹⁾ gas core reactors are ideal irradiators because they provide an abundant supply of neutrons for transmutation.

An interesting characteristic of externally moderated reactors is that the effective neutron temperature of the thermal flux is determined by the temperature of the external moderator. Hence, by controlling the temperature of the beryllium reflector, reactivity control of the core can be affected.

Actinide Blanket Design Considerations

Liquid metal fuel reactor systems have been studied extensively.⁽¹²⁾ The present liquid bismuth-actinide blanket design relied substantially on information gathered in these early works.

In this design, the blanket consists of 93 wt% liquid bismuth and 7 wt% actinides. Since solubilities of actinide metals in liquid bismuth are not well established, the blanket may take the form of a homogeneous solution or that of an actinide-bismuth slurry. For the case of a slurry, the actinides are present as small particles dispersed uniformly throughout the bismuth. Additional attention will have to be directed towards the problems of concentration control, stability, and erosion.

The use of a liquid bismuth-actinide blanket has many advantages. Continuous reprocessing of fission product poisons can be carried out to ensure maximum utilization of neutrons for transmutation. Continuous refueling of the blanket leads to great flexibility in actinide fuel management. Since the fluid fuel can be cooled in an external heat exchanger separate from the reactor core, the nuclear requirements (of the core) and heat flow requirements (of the exchanger) need not both be satisfied at the same place. This may allow design for very high specific power. Furthermore, liquid bismuth can be operated at high temperatures without high pressures, is free from radiation damage, and has better heat transfer properties than water.

Bismuth is quite corrosive to most metals and alloys,⁽¹³⁾ but its corrosiveness can be reduced (particularly with respect to steel) by the addition to the bismuth of zirconium or titanium in conjunction with magnesium. The zirconium (or titanium) is believed to react with nitrogen and/or carbon in the steel to form a protective layer of ZrN or ZrC which provides a barrier between the bismuth and the ferrous alloy substrate.^(14,15) The role of magnesium in conjunction with zirconium or titanium is to getter oxygen from the system, thereby preventing any oxidation of the latter two elements which would destroy their effectiveness.⁽¹⁶⁾ The materials that can be used to contain bismuth-uranium fuels are graphite, beryllium, carbon steels, low chromium steels, molybdenum, tantalum, tungsten, and some high alloy steels.⁽¹⁶⁾

A 20 cm thickness of blanket is chosen. Assuming a maximum volumetric heat generation rate of 200 watts/cc, the maximum blanket power is about 1600 MWt. No detailed analysis of heat transfer and fluid flow of the

TABLE 3.3

Core Neutronic Parameters for Different Blanket Composition

Core power = 2000 MWt

Bi = U = 93 = 7 Wt%

$\frac{U^{233}}{U^{238}}$ ratio	$\frac{0}{100}$	$\frac{0.0625}{99.9375}$	$\frac{0.125}{99.875}$	$\frac{0.25}{99.75}$
Blanket Power (MWt)	~ 0	180	370	790
Core peak-to-average power ratio	1.002	1.002	1.002	1.002
Core critical mass (kg of U^{233})	6.57	6.45	6.32	6.05

blanket was performed. However, no major difficulties are anticipated. An average blanket temperature of 723°K (450°C, 842°F) was assumed.

UFATR Core Neutronic Characteristics

Twenty five-group MACH-1 calculations are performed for the UFATR. Thermal neutrons induce fission in the gas core, leading to the production of 2.5 fast neutrons per fission. These fast neutrons quickly escape to the beryllium moderator and are thermalized. Some of them are returned to the core to maintain the self-sustaining reaction. A substantial portion (35%-50%) are trapped in the bismuth-actinide blanket leading to transmutation reactions. Figure 3.5 is a plot of neutron flux integrals in the core as a function of energy groups. Since a 25-group actinide cross section set was not available, their presence was simulated by a U^{233} - U^{238} mixture. The U^{233} fraction in the blanket was varied so that a blanket power from 0 to 800 MWt was produced. This would simulate the changing neutronic characteristics of the actinide blanket during irradiation. Table 3.3 shows some core parameters for different blanket compositions. Two desirable characteristics of the UFATR can be observed. The peak-to-average ratio of the core power is extremely close to unity. The core parameters are insensitive to changes in the actinide blanket composition.

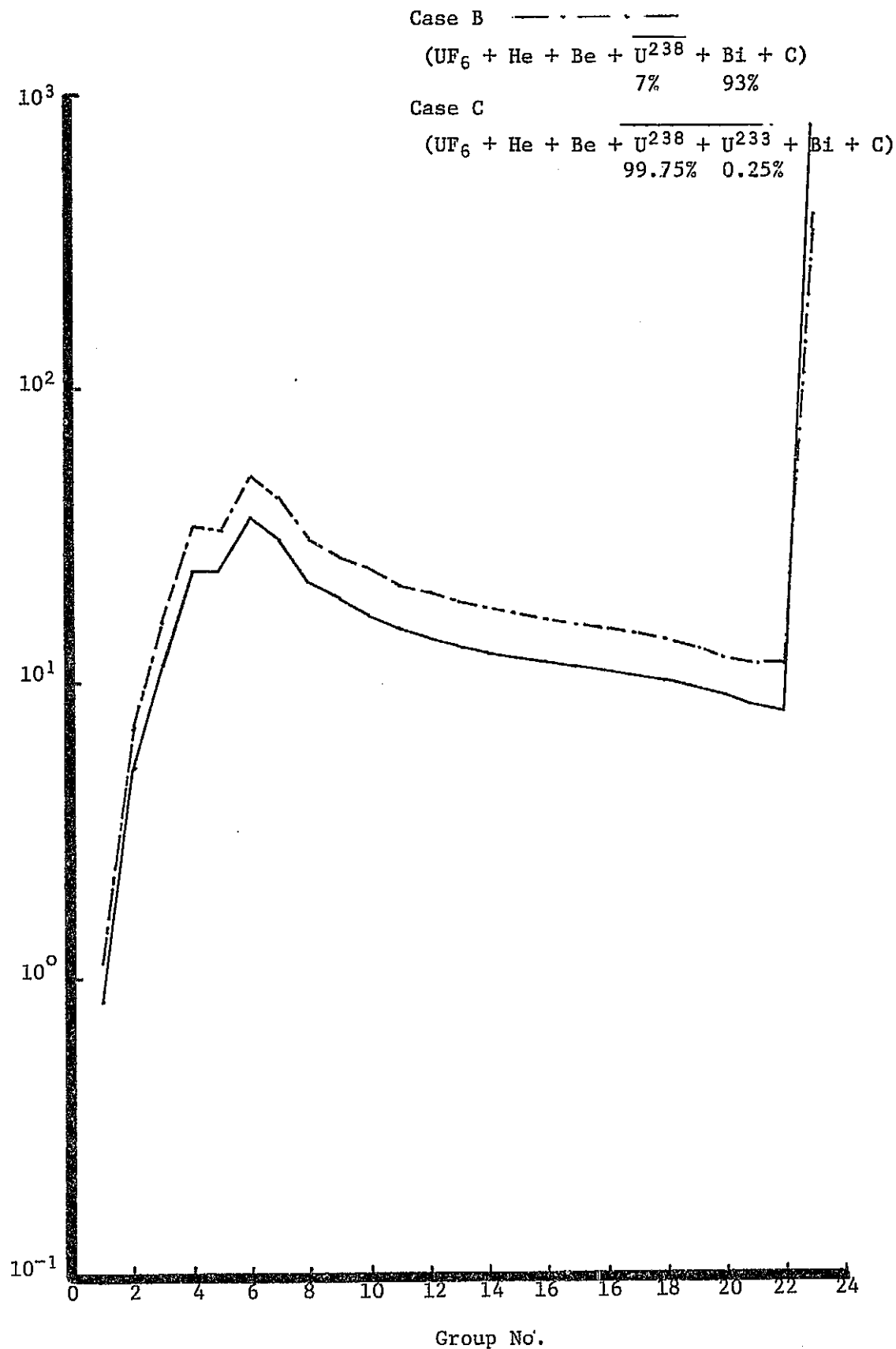


Fig. 3.5. Flux Integral vs Energy Group in $\text{UF}_6 + \text{HE}$ Core Region

References for Chapter 3

1. Meneley, D. A., Kvitek, L. C. and O'Shea, D. M., "MACH-1, A One-Dimensional Diffusion-Theory Package," ANL - 7223 (June, 1966).
2. Abagyan, L. P., Bazazyants, N. O., Bondarenko, I. I. and Nikolaev, M. N., Group Constants for Nuclear Reactor Calculations, Consultants Bureau, New York (1964).
3. Mills, C. B., "Reactor Minimum Critical Dimensions," LA-3221-MS (April, 1965).
4. Mills, C. B., "Reflector Moderated Reactors," Nuclear Science and Engineering, 13, 301-305 (1962).
5. Westcott, C. H., "Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra," CRRP-787 (August, 1958).
6. Garber, D. I. and Kinsey, R.R., "Neutron Cross Sections," 3rd Edition, Vol II, BNL-325 (January, 1976).
7. Hughes, D. J. and Schwartz, R. B., "Neutron Cross Sections," 2nd Edition, BNL-325 (July, 1958).
8. Clement, J. D. and Rust, J. H., "Gas Core Reactors for Actinide Transmutation and Breeder Applications," NASA Grant NSG-1288 (April, 1978).
9. Wagner, P., "Materials Considerations for UF_6 Gas-Core Reactor," LA-6776 - MS (April, 1977).
10. O'Donnell, P. M., "Kinetics of the Fluorination of Beryllium," J. Electrochem. Soc., 114, 1206-09 (1967).
11. Safonov, G., "Externally Moderated Reactors," Proc. Intern. Conf. Peaceful Uses of Atomic Energy, Geneva, 12, 705 (1958).
12. Lane, E. J. MacPherson, H. G. and Maslan, F., Ed., Fluid Fuel Reactors, Addison-Wesley Publisher Company, Inc. (September, 1958).
13. Berry, E. W., Corrosion in Nuclear Applications, John Wiley and Sons, Inc. (1971).
14. Romano, A. J., Klamut, C. J. and Gurinsky, D. H., "The Investigation of Container Materials for Bi and Pb Alloys, Part I, Thermal Convection Loops," BNL-811 (July, 1963).
15. Kammerer, O. F., Weeks, J. R., Sadofsky, J., Miller, W.E. and Gurinsky, D. H., "Zirconium and Titanium Inhibit Corrosion and Mass Transfer of Steels by Liquid Heavy Metals," Trans. Met. Soc. AIME, 212 (1), 20-25 (February, 1958).
16. Klamut, C. J., Schweitzer, D. G., Chow, J.G.Y., Meyer, R.A., Kammerer, O.F., Weeks, J. R. and Gurinsky, D. H., "Material and Fuel Technology for an LMFBR," Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 7, 173-195 (September, 1958).

Chapter 4.

ANALYSIS OF ACTINIDE BURNUP IN UFATR

Actinide Cross Sections

The validity of actinide transmutation calculations are dependent upon the accuracy of actinide neutron cross sections. A large number of reactor concepts, including LWRs, LMFBRs, CTRs have been studied as transmutation candidates. Therefore, the range over which capture and fission cross sections of actinides need to be known extends from thermal to about 18 MeV of neutron energy.⁽¹⁻⁶⁾ There are 16 trans-actinium elements with 200 isotopes known to date. For many of these actinides, experimental data may not exist. This is due to short half lives, an inability to obtain samples of sufficient isotopic purity, and difficulty of obtaining higher energy (are 14 MeV) nonenergetic neutron sources for differential cross section measurement. Consequently, for many of these, the necessary data has been obtained by application of nuclear systematics and model calculations.⁽⁷⁾ Generally, the main isotopes of Th, Pa, U, Np, and Pu have been evaluated extensively. There is an urgent need for evaluation of americium and curium isotopes cross sections, and to a lesser extent, those of berkelium and californium. For higher actinide isotopes, they usually are very short lived and exist in such minute quantities that they are insignificant for most applications. The thermal cross sections of the actinides have been found to yield computational results in agreement with experimental data from transplutonium production programs.⁽⁸⁻¹²⁾ As one moves away from the thermal region into the fast energy region, greater uncertainty persists.

For the present calculation, a three-group actinide cross section set was generated as shown in Table 4.1. Only those nuclides which may

TABLE 4.1

Three Group Cross Section Set for the Actinides

Group # Nuclide	1 (FAST)			2 (RESONANCE)			3 (THERMAL)		
	σ_a	$\nu\sigma_f$	ν	σ_a	$\nu\sigma_f$	ν	σ_a	$\nu\sigma_f$	ν
U ²³²	*2.63-3	0.0	0.0	42.0	70.1	3.13	2.63-3	0	0.0
U ²³⁴	1.93	5.06	2.62	44.1	0.0	0.0	54.56	0.0	0.0
U ²³⁶	1.62	4.31	2.66	25.55	0.0	0.0	2.83	0.0	0.0
U ²³⁷	1.88	5.19	2.76	84.0	0.0	0.0	205.85	0.0	0.0
Np ²³⁷	1.22	3.60	2.95	4.62	0.0	0.0	103.08	2.76-2	2.67
Np ²³⁸	0.0	0.0	0.0	61.6	17.06	2.77	1127.0	3123.0	2.77
Pu ²³⁶	0.0	0.0	0.0	0.0	0.0	0.0	88.2	244.4	2.77
Pu ²³⁷	0.0	0.0	0.0	0.0	0.0	0.0	154.0	440.4	2.86
Pu ²³⁸	2.63-3	0.0	0.0	13.23	5.07	2.895	313.8	27.27	2.895
Pu ²³⁹	2.06	6.4	3.2	35.07	21.07	2.87	1129.3	2080.7	2.87
Pu ²⁴⁰	1.23	3.825	3.11	560.9	0.0	0.0	185.2	8.37-2	2.79
Pu ²⁴¹	7.90-3	0.0	0.0	51.24	116.67	2.924	1099.3	2369.4	2.924
Pu ²⁴²	7.40-3	0.0	0.0	89.58	0.925	2.81	10.44	0.0	0.0
Pu ²⁴³	1.29	4.22	3.27	56.42	110.4	2.91	14.56	285.25	2.91
Pu ²⁴⁴	0.0	0.0	0.0	2.975	0.0	0.0	0.926	0.0	0.0
Am ²⁴¹	1.10	0.0	3.41	81.97	4.59	3.121	578.4	8.76	3.121
Am ²⁴²	1.85-2	0.0	0.0	0.0	0.0	0.0	1143.6	3636.7	3.18
Am ^{242m}	1.83	6.44	3.52	109.9	358.7	3.264	5227.9	1.351+4	3.26
Am ²⁴³	0.0	0.0	0.0	135.1	0.7224	3.09	28.1	0.0	3.09
Cm ²⁴²	0.0	0.0	0.0	10.50	0.0	3.19	13.61	8.686	3.19
Cm ²⁴³	5.0-3	0.0	0.0	165.2	446.6	3.43	484.7	1.289+3	3.43
Cm ²⁴⁴	0.0	0.0	0.0	42.2	4.01	3.20	6.37	1.917	3.20
Cm ²⁴⁵	0.0	0.0	0.0	60.9	205.48	3.832	1.385+3	4.5096+3	3.832
Cm ²⁴⁶	0.0	0.0	0.0	8.886	2.644	3.80	0.8768	0.3518	3.80
Cm ²⁴⁷	0.0	0.0	0.0	88.3	202.4	3.80	70.96	149.2	3.79
Cm ²⁴⁸	0.0	0.0	0.0	18.59	4.013	3.90	1.759	0.7221	3.90

* read as 2.63×10^{-3}

have a significant effect on the blanket neutronics are included. Whenever possible, the more up-to-date data of Benjamin⁽¹¹⁾ is used to supplement the ORIGEN data library.⁽¹³⁾ The cross sections are spectrum-averaged. The fast energy group extends from 10 MeV to 0.8 MeV. A fission neutron spectrum is assumed for this region. The resonance region extends from 0.8 MeV to 0.465 eV. A $\frac{1}{E}$ spectrum is assumed. The thermal region extends from 0 eV to 0.465 eV with a Maxwellian spectrum assumed. Cross sections of Np^{237} , Pu^{239} , Pu^{240} , Pu^{241} , Pu^{242} , Am^{241} , and Am^{243} are corrected with non- $1/v$ factors from Westcott.⁽¹⁴⁾ For the other nuclides, $1/v$ behavior of cross sections is assumed. The downscattering cross sections for the actinides are approximated by those of U^{238} . Since the actinides are heavy nuclides and present in low concentration, they should have little effect on the neutron transport characteristics of the liquid bismuth blanket.

Computational Strategy

Figure 4.1 illustrates the computational strategy used for analyzing the actinide blanket as a function of burnup. MACH-1 is used for neutronic analysis. The 26-group ABBN cross section set is collapsed to a 3-group cross section set for use in conjunction with the actinide cross sections generated previously. The code ORIGEN⁽¹³⁾ is used to keep track of buildup and depletion of actinides during irradiation. The concentrations of actinides are inputted into MACH-1, which calculates the neutron flux distribution in the reactor. This information is used to generate the parameters, THERM, RES, FAST, and FLUX that are required for ORIGEN input. ORIGEN then calculates actinide concentrations at the end of the time step. This procedure is repeated.

Actinide Fuel Management During Burnup

Numerous actinides fuel management schemes are possible during irradiation of actinides. Detailed studies are required to choose a management scheme that will maximize the actinide burnup performance. For the present study, the following strategy is used. At the beginning of life, 6.0 metric tonnes of actinides is charged into the blanket. This quantity of actinides is equivalent to the amount produced from 300 LWR-years of operation. The reactor is operated with a core power of 2000 MWt. Since the blanket is very subcritical, only a small amount of power is produced. The liquid bismuth blanket is circulated to remove heat produced and for reprocessing of fission products. No refueling of actinides is carried out. As actinide nuclides are converted to more fissile isotopes, the blanket power rises. Eventually, it will reach 1600 MWt — the maximum design power for the blanket. At this point in time, continuous refueling is introduced. The addition of the poor quality actinide feed makes the blanket become more subcritical. The blanket is operated at a constant power of 1600 MWt. The actinide refueling rate is set to match the depletion rate so the actinide inventory in the blanket is maintained constant. The blanket composition will change with time until equilibrium is reached.

Berwald⁽¹⁵⁾ found a problem in the ORIGEN code when the continuous refueling option is chosen. His prescription for the correction of this error was adopted.

Analysis of Actinide Burnup Performance

Table 4.2 shows the core and blanket neutronic parameters as a function of burnup. It should be noted that core critical mass stays relatively constant for the 10 year irradiation period. For the first 4 years, the

TABLE 4.2

Core and Blanket Parameters as a Function of Burnup

Time		0	1Y	2Y	3Y	4Y	5Y	6Y	7Y	8Y	10Y
Core	U^{233} mass (kg)	7.05	6.83	6.74	6.53	6.30	6.23	6.21	6.21	6.21	6.21
	flux	$1.16+16$	$1.20+16$	$1.21+16$	$1.25+16$	$1.30+16$	$9.29+15$	$9.16+15$	$9.17+15$	$9.22+15$	$9.03+15$
	power	2000	2000	2000	2000	2000	1410	1390	1390	1400	1370
Blanket	flux	$4.11+13$	$5.67+13$	$5.16+13$	$5.41+13$	$4.88+13$	$4.78+13$	$4.82+13$	$4.90+13$	$4.98+13$	$5.02+13$
	power	239	702	1109	1591	1600	1600	1600	1600	1600	1600

core power is set at 2000 MWt. During this time, the blanket is becoming more fissile and its power is rising rapidly. At the end of 4 years, the blanket power is approaching the design maximum of 1600 MWt. At this point, continuous refueling of actinides is carried out with the feed rate equalling the depletion rate. The blanket power is held constant at 1600 MWt; however, the core power is dropped to 1400 MWt. At the end of 10 years, the blanket is very close to equilibrium. Figure 4.2 shows the blanket power and flux as a function of burnup.

Table 4.3 shows the quantity of the more abundant actinides in the blanket during burnup. An initial load of 6 metric tonnes of actinides is charged. At the end of 4 years, the inventory is reduced to 3.9 metric tonnes. From 4 to 10 years, the blanket composition stabilizes very quickly, and is close to equilibrium after 10 years.

Table 4.4 shows the principal fissioning nuclides in the blanket as a function of time. At the beginning of life, the power is mostly coming from $\text{Am}^{242\text{m}}$, Cm^{245} and Np^{237} . As the irradiation proceeds, the blanket becomes more fissile due to the accumulation of plutonium isotopes. At the end of 10 years, the principal fissioning nuclides are Pu^{239} , Cm^{245} , and Pu^{241} . This change in blanket composition and criticality is reflected in changes in values of RES and FAST, as shown in Table 4.5. The parameters RES and FAST are proportional to the neutron flux in the resonance and fast regions, respectively, relative to the thermal flux. Figure 4.3 is a plot of the blanket fission densities as a function of radial distance for different burnup times.

To further evaluate the criticality of the blanket during burnup, MACH-1 analysis of the blanket is performed. The fissioning gas core is replaced by solid beryllium. The effective multiplication constant of the

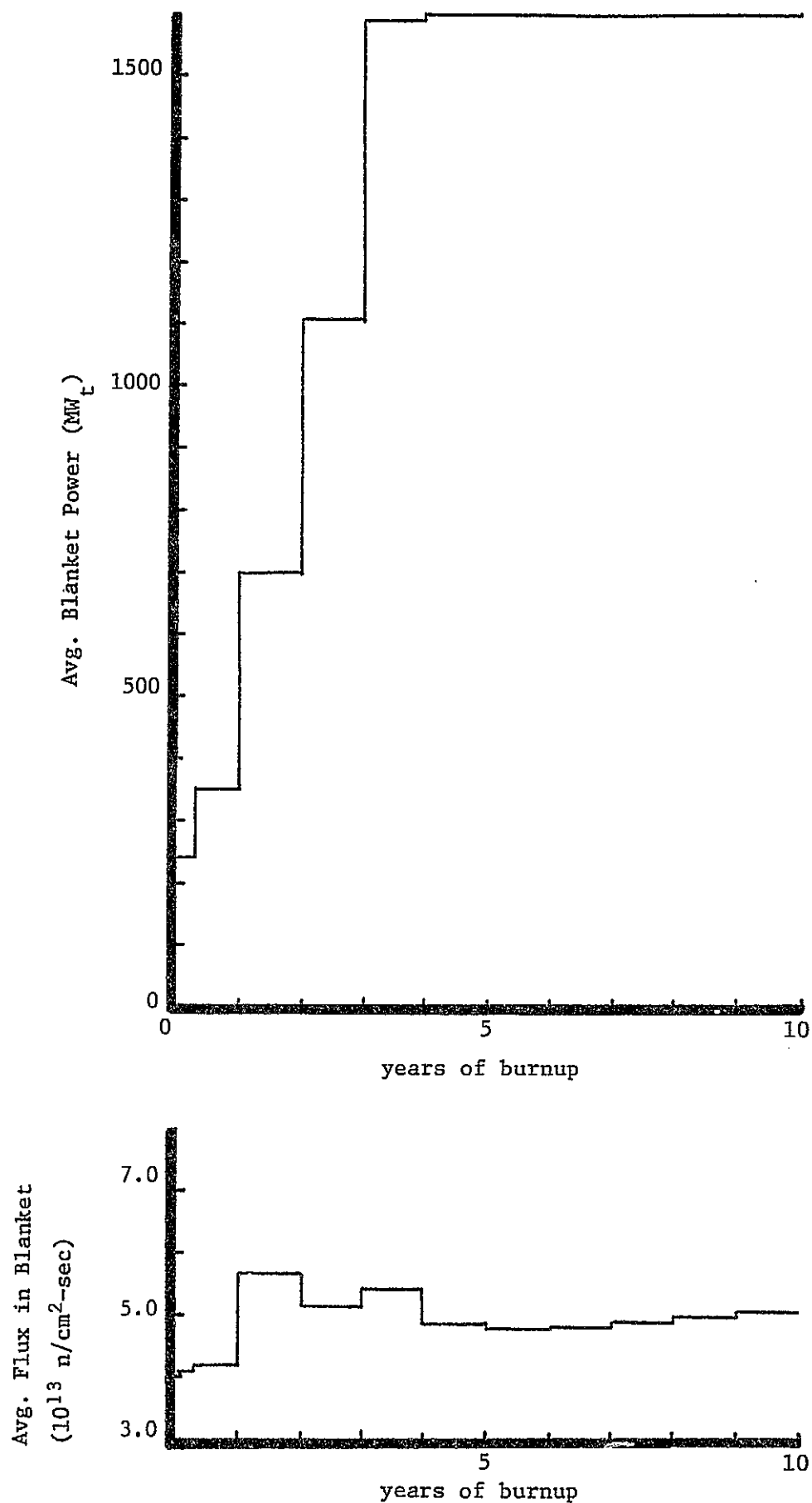


Fig. 4.2. Plot of Avg. Flux and Power in Blanket as a Function of Burnup Time

TABLE 4.3

Gm-Atoms of the Principal Actinides as a Function of Burnup

Nuclide	Charge	1Y	2Y	3Y	4Y	5Y	6Y	7Y	8Y	10Y
Np ²³⁷	1.85+4	1.52+4	1.13+4	8.23+3	5.56+3	5.39+3	5.29+3	5.21+3	5.15+3	5.04+3
Np ²³⁸		2.37+1	2.70+1	2.07+1	1.71+1	1.59+1	1.57+1	1.56+1	1.55+1	1.54+1
Pu ²³⁸		3.00+3	3.36+3	4.42+3	4.49+3	4.29+3	4.18+3	4.10+3	4.03+3	3.90+3
Pu ²³⁹		3.90+2	5.89+2	9.43+2	1.07+3	1.05+3	1.02+3	9.95+2	9.75+2	9.42+2
Pu ²⁴⁰		1.10+2	3.08+2	5.56+2	7.19+2	7.60+2	7.61+2	7.53+2	7.46+2	7.32+2
Pu ²⁴¹		1.21+1	6.69+1	1.54+2	2.64+2	3.24+2	3.39+2	3.38+2	3.32+2	3.23+2
Pu ²⁴²		1.52+2	2.09+2	2.30+2	2.68+2	3.26+2	3.85+2	4.31+2	4.63+2	4.97+2
Am ²⁴¹	1.75+3	7.05+2	1.96+2	5.72+1	1.83+1	1.09+2	1.32+2	1.37+2	1.37+2	1.35+2
Am ²⁴²	4.22-4	1.34+0	5.49-1	1.60-1	5.98-2	3.37-1	4.08-1	4.27-1	4.31-1	4.31-1
Am ^{242m}	3.52+1	1.90+1	5.93+0	1.91+0	6.47-1	3.88+0	4.85+0	5.05+0	5.03+0	4.91+0
Am ²⁴³	3.53+3	2.92+3	2.12+3	1.49+3	9.61+2	9.51+2	9.67+2	9.92+2	1.02+3	1.05+3
Cm ²⁴²	4.09+1	3.27+2	2.13+2	8.52+1	3.17+1	4.95+1	7.28+1	8.28+1	8.62+1	8.72+1
Cm ²⁴³	2.93+0	5.92+0	6.65+0	4.03+0	1.69+0	1.13+0	1.36+0	1.62+0	1.77+0	1.87+0
Cm ²⁴⁴	1.06+3	1.60+3	2.17+3	2.45+3	2.49+3	2.50+3	2.51+3	2.53+3	2.56+3	2.63+3
Cm ²⁴⁵	7.17+1	5.55+1	9.51+1	1.45+2	1.97+2	2.25+2	2.32+2	2.33+2	2.32+2	2.35+2
Cm ²⁴⁶	8.18+0	2.41+1	5.16+1	9.24+1	1.54+2	2.20+2	2.87+2	3.53+2	4.17+2	5.42+2
Totals	2.50+4	2.46+4	2.07+4	1.90+4	1.64+4	1.63+4	1.64+4	1.63+4	1.63+4	1.64+4

TABLE 4.4

Percentage of Blanket Power from Actinides as a Function of Burnup

Nuclide	0 days	1Y	2Y	3Y	4Y	5Y	6Y	7Y	8Y	10Y
Np ²³⁷	12.7	9.3	7.0	5.0	3.4	3.3	3.3	3.2	3.2	3.2
Np ²³⁸	2.4	5.1	4.2	2.2	1.5	1.3	1.3	1.3	1.3	1.3
Pu ²³⁸		3.9	5.3	4.9	4.3	3.9	3.8	3.7	3.7	3.7
Pu ²³⁹		51.6	55.6	58.7	55.2	51.0	49.5	48.9	48.8	48.2
Pu ²⁴¹		1.9	7.5	11.5	16.4	19.0	19.8	20.0	20.0	19.9
Am ^{242m}	52.6	14.5	3.3			1.1	1.4	1.5	1.5	1.5
Cm ²⁴⁵	30.9	12.2	15.3	15.6	17.7	19.0	19.6	19.9	20.1	20.8

TABLE 4.5

Parameters RES and FAST as a Function of Burnup

Time	0 days	1Y	2Y	3Y	4Y	5Y	6Y	7Y	8Y	10Y
RES	0.0533	0.0888	0.121	0.163	0.186	0.189	0.188	0.185	0.182	0.178
FAST	2.29	4.04	5.79	8.82	10.79	11.52	11.56	11.42	11.24	11.01

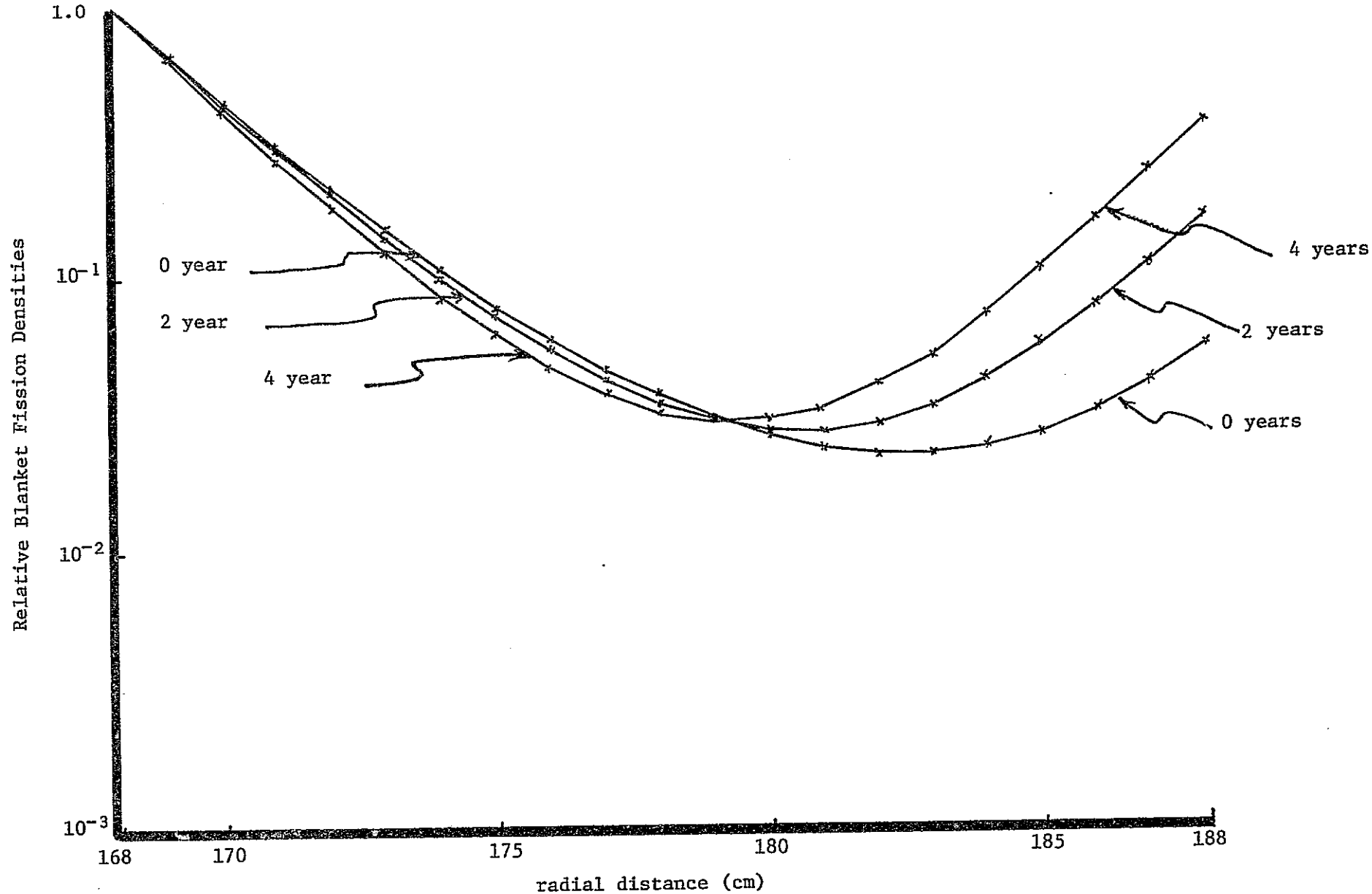


Fig. 4.3. Relative Blanket Fission Densities as a Function of Radial Distance at Different Times

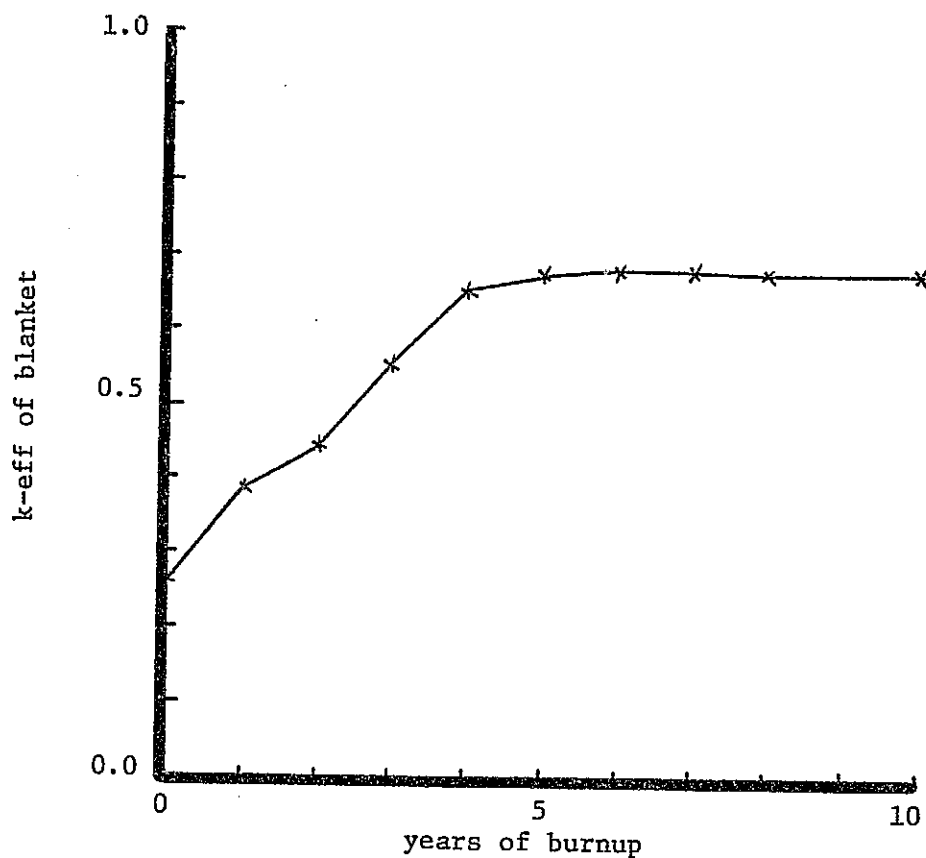


Fig. 4.4. Effective Multiplication of Blanket as a Function of Burnup Time.

blanket is calculated by MACH-1. Figure 4.4 is a plot of k_{eff} as a function of burnup time. At the beginning of life, the blanket is grossly subcritical with an effective multiplication constant of 0.26. At 5 years, the effective multiplication constant has risen to 0.68. At this point, blanket power is 1600 MWt and blanket criticality is suppressed by the addition of poor quality fresh actinide feed. It is conceivable that the blanket may become critical by itself.

Actinide Production in Core

A basic requirement of all transmutation schemes is that the quantity of actinides produced by the transmutation system must be less than the quantity of actinides that are destroyed. For the gas core reactor systems, actinides are produced by the capture reaction of U^{233} . Table 4.6 shows the capture-to-fission ratio as a function burnup. Taking a maximum capture-to-fission ratio of 0.013, and a core power of 2000 MWt, actinides production rate is 8.11×10^{17} atoms/sec. To take care of this quantity of actinides, assuming 200 MeV per fission, a power of 26 MWt is required. That is, the blanket power must be greater than 26 MWt in order for the reactor system to destroy actinides at a rate higher than their production rate in the core. For the UFATR, this requirement is met with little difficulty.

TABLE 4.6

Alpha (capture-to-fission ratio) of U^{233} as a Function of
Actinide Burnup.

Time	0 Y	1 Y	2 Y	3 Y	4 Y	5 Y	6 Y	7 Y	8 Y	10 Y
Alpha	0.013	0.012	0.012	0.012	0.012	0.011	0.011	0.011	0.011	0.011

References for Chapter 4

1. Mann, F. M. and Schenter, R. E., "Actinide Cross-Section Calculations and Evaluations," Trans. Am. Nucl. Soc., 23, 546-547 (1976).
2. Morrison, G. W., Burns, T. J. and Weisbin, "Actinide Transmutation: Cross Sections Methods, and Reactor Sensitivity Analysis," Trans. Am. Nucl. Soc., 23, 552 (1976),
3. Raman, S., "General Survey of Applications Which Require Actinide Nuclear Data," Proc. IAEA Advisory Group Mtg. Transactinium Isotope Nuclear Data, CONF - 751104 - 4, Review Paper A1 (1975).
4. Kusters, H. and Lalovic, M., "Review of Transactinium Isotope Build-Up and Decay in Reactor Fuel and Related Sensitivities to Cross-Section Changes and Results and Main Conclusions of the IAEA - Advisory Group Mtg. on Transactinium Nuclear Data Held at Karlsruhe, November, 1975," KFK-2283 (1976).
5. Yiftah, S., Gur, Y. and Caner, M., "Status of Transactinium Isotope Evaluated Nuclear Data in the Energy Range 10^{-3} eV to 15 MeV," IAEA-186, 165-194 (1976).
6. Dabbs, J. W. T., "The Nuclear Fuel Cycle and Actinide Wastes: Cross Section Needs and Recent Measurements," ORNL/TM - 5530 (1976).
7. Mann, F. M. and Schenter, R. E., "Actinide Cross Section Calculations and Evaluations," HEDL-SA-814 (1976).
8. Graves, W. E. and Benjamin, R. W., "Predicting Production Rates of Heavy Actinides," CONF-720901, 97-107 (1972).
9. Hennelly, E. J., "The Heavy Actinide Cross Section Story," CONF-710301, 2, 494-501.
10. Benjamin, R. W., McCrosson, F. J., Gorrell, T. C. and Vandervelde, V. D., "A Consistent Set of Heavy Actinide Multigroup Cross Sections," DP-1394 (1975).
11. Benjamin, R. W., "Status of Measured Neutron Cross sections of Transactinium Isotopes for Thermal Reactors," DP-MS-75-87 (1975).
12. Hennelley, E. J., "Nuclear Data for Actinide Recycle," Natl. Bur. Stands. Spec. Publ., CONF-750303-P1 (Oct., 1975).
13. Bell, M. J., "ORIGEN - The ORNL Isotope Generations and Depletion Code," ORNL-4628 (May, 1973).
14. Westcott, C. H., "Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra," CRRP-787 (August, 1958).
15. Berwald, D. H., "Preliminary Design and Neutronic Analysis of a Laser Fusion Driven Actinide Waste Burning Hybrid Reactor," Ph. D. Thesis, Dept. of Nuclear Engineering, U. of Michigan (1977).

CONCLUSIONS AND RECOMMENDATIONS

Preliminary design and analysis of a uranium hexafluoride actinide transmutation reactor (UFATR) is performed. The purpose of this reactor is to convert long-lived actinide wastes to shorter-lived fission product wastes.

It is demonstrated that externally moderated gas core reactors are ideal irradiators. They provide an abundant supply of thermal neutrons and are insensitive to composition changes in the blanket.

For the present UFATR, an initial load of 6 metric tonnes of actinides is loaded. This is equivalent to the quantity produced by 300 LWR-years of operation. Initially, the core produces 2000 MWt and the blanket 239 MWt. After 4 years of irradiation, the actinide mass is reduced to 3.9 metric tonnes. With continuous actinide refueling, the actinide inventory is held constant and equilibrium essentially achieved at the end of 8 years. At equilibrium, the core produces about 1400 MWt and the blanket 1600 MWt. At this power level, the actinide destruction rate is equal to the production rate from 32 LWRs.

Recommendations

To further evaluate the UFATR, more design and research work is required in several areas. To be able to transmute actinides effectively, they must be extracted from bulk waste at high efficiencies. Research work on the chemical reprocessing of actinides is needed. The accuracy of actinide depletion calculations is strongly dependent on the precision of actinide cross sections. In particular, the americium and curium cross sections are very significant in determining blanket neutronic

characteristics. Detailed differential cross section measurements are required. The actinide fuel management strategy adopted for the present study is one of many possible ones. Future work should concentrate on defining a strategy that will optimize actinide burnup performance. For the present design, a maximum blanket effective multiplication constant of 0.68 is attained. It is conceivable that the blanket can become critical by itself. To complete the system design of the UFATR, more work is required in the areas of heat transfer and fluid flow of the UF_6 -He fuel and liquid bismuth-actinide solution (or slurry). Additional work is required in the continuous reprocessing and refueling of the UF_6 -He fuel and liquid bismuth-actinide solution (or slurry).

Appendix A Reprocessing Systems

No quantitative analysis was made of the reprocessing systems for the UF_6 breeder and actinide transmutation reactors. However, since the reprocessing systems are important to the operation of the power plants, a qualitative discussion is included in this study which is based on proposed systems given in Refs. 1 and 2. Although these studies were preliminary in nature, they did not encounter major obstacles.

There are two major reprocessing systems to be considered. The first is the cleanup of fission products in the UF_6 -helium mixture. The second is the extraction of the actinides from other waste products to be used in the actinide transmutation reactor. These systems will be described in the following sections.

A.1 Fission Product Cleanup

Fission products must be removed from the UF_6 -helium mixture continuously to avoid buildup of reactor poisons and condensation of volatiles. Fortunately, the technology for UF_6 separation and purification is available from the Molten Salt Breeder Reactor Program at Oak Ridge National Laboratory and helium purification technology is available from the High Temperature Gas Cooled Reactor developed by General Atomics.

It is expected that some UF_6 will dissociate in the core and that the fluorine formed will combine with metallic fission products to form fluorides. According to Ref. 1, the fluorides and gases in Table A.1 will be formed. The fluorides are divided into volatile, mobile, intermediate and refractory fluorides according to their boiling points. The mole fractions of the fission product gases, volatile fluorides, and mobile fluorides are on the order of 10^{-5} less than the mole fraction

TABLE A.1
Gaseous and Fluoride Fission Products⁽¹⁾

Gases	Volatile Fluorides	Mobile Fluorides	Intermediate Fluorides	Refractory Fluorides
Kr	Se F ₆ (236°K)*	Sb F ₅ (423°K)	Cs F (1524°K)	Ra F ₂ (2410°K)
Xe	Mo F ₆ (308°K)	Nb F ₅ (509°K)	Rb F (1663°K)	Y F ₃ (2500°K)
I	Te F ₆ (309°K)	Ru F ₅ (523°K)		Ce F ₃ (2573°K)
Br		Zr F ₅ (873°K)		Nd F ₃ (2573°K)
		Su F ₄ (978°K)		Pr F ₄ (2600°K)
				La F ₃ (2600°K)
				Sr F ₂ (2762°K)

* numbers in parantheses are the boiling points of the various fluorides

of helium while the mole fractions of the intermediate and refractory fluorides are 10^{-3} less than the other fluorides.

Due to their low boiling points, the volatile and some of the mobile fluorides will remain in the UF_6 -helium circulating gas loop until they are removed for reprocessing. The other fluorides will be deposited in the heat exchangers and piping. The problem is further complicated by radioactive decay of various species, resulting in a change of their chemical nature and the relocation of their deposition sites.

Reference 1 suggests that replaceable getter pads made of nickel wire be placed in the reactor outlet piping to capture the intermediate and refractory fluorides.

Lowry⁽¹⁾ of the Los Alamos Scientific Laboratory proposed the fission product cleanup system shown in Fig. A.1. A small amount of UF_6 -helium gas mixture is bled from the circulating loop and is reduced in pressure to 1.5 atmospheres. The mixture then passes into a high temperature bed of NaF pellets at 500°K where most of the volatile fluorides are absorbed and is cooled to 300°K before entering a low temperature bed of NaF pellets. The low temperature bed absorbs the UF_6 and remaining metal fluorides while the helium containing xenon, krypton, bromine, iodine and other gases pass through the filter to the helium purification system.

Two low temperature beds are utilized. When one bed becomes loaded with UF_6 , the flow into this bed is valved out and the fresh bed is placed in service. The bed loaded with UF_6 is then heated to 700°K which drives off UF_6 as a gas along with small amounts of TeF_6 . A helium purge gas is used to help remove the UF_6 . Finally, the UF_6 passes through a

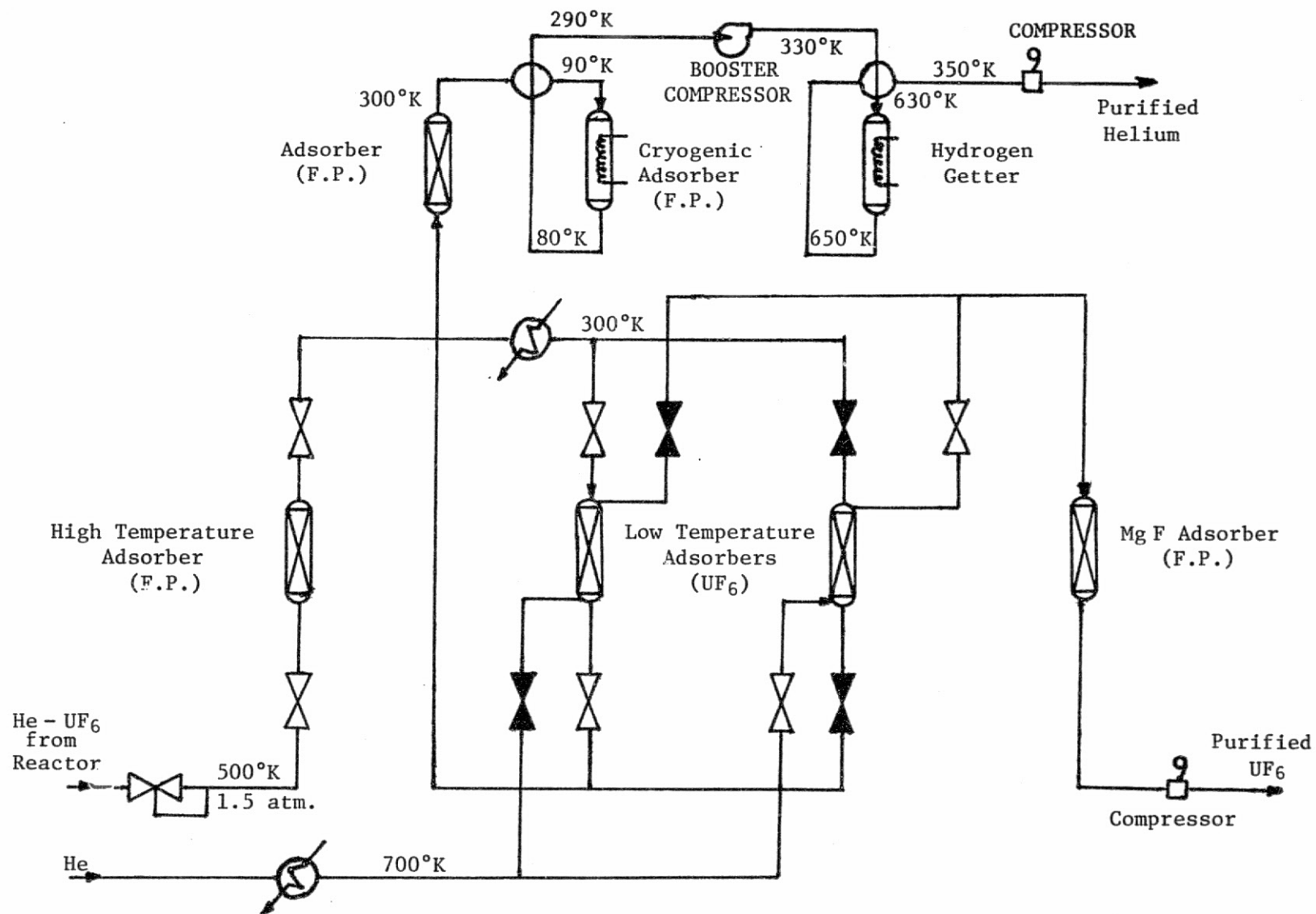


Fig. A.1 Fission Product Removal System⁽¹⁾

bed of MgF_2 to remove the TeF_6 before being filtered, pressurized, and cooled to produce a purified liquid which is recycled to the reactor. The NaF and MgF_2 beds containing fission products are either stored or sent to a waste treatment plant.

Helium at 300°K flows into one of two parallel systems consisting of high and low temperature charcoal absorbers. The high temperature absorber contains activated charcoal impregnated with potassium. The charcoal removes the condensable metallic fission products while the potassium removes iodine by chemisorption.

The helium is then cooled to 90°K in a helium regenerator and passes through the low temperature absorber which removes krypton, xenon, nitrogen, and some hydrogen and tritium. Helium is cooled in the absorber to 80°K by liquid nitrogen. The purified helium then enters the cold side of the regenerator where it is heated to 290°K and is filtered to remove dust before being compressed and sent to the hydrogen removal section.

Helium leaving the compressor enters another regenerator before passing through one of two parallel hydrogen getters consisting of titanium sponges to remove hydrogen and tritium. Helium enters the getters at 630°K and is heated by the electrically heated sponges to 650°K . The helium then reenters the regenerator and is cooled to 350°K , filtered and recompressed.

The uranium inventory in the reprocessing system is not a function of reactor power but of regeneration frequency and volume of the NaF bed.

Distillation⁽¹⁾ is an alternative method for fission product removal especially if a large part of the primary stream must be cleaned up. The bled stream enters a distillation column where most of the fluorides are

removed as a concentrate at the bottom of the column. An aqueous wash removes the fluorides from the concentrate and residual UF_6 is returned to the column for further purification. The UF_6 and volatile fluorides are condensed and fed to a second column which produces pure UF_6 at the bottom of the column.

Another method for UF_6 purification is a combination of a cold trap process and fluoride volatility process proposed by Rust and Clement.⁽²⁾

Clearly, there are several possible methods for UF_6 purification. The method that will be selected should be based on consideration of economics, minimum uranium inventory, effectiveness in keeping the system as clean as possible, and compatibility with power plant operation.

A.2 Actinide Reprocessing System

Because of the hazardous radionuclides present in high-level wastes from present day reactors, schemes are needed which provide waste management programs of one million years or longer.

One alternative to this would be to remove the long-lived actinides which require long term surveillance. If this could be achieved, the remaining fission products and wastes would require a waste management program on the order of 1000 years. The actinides would then be transmuted in a fission or other type reactor to reduce the long half-lives to short ones, and thus reduce the radioactive hazard. The main problem to be overcome is separation of actinides from the rest of the waste products.

With the assumption that this separation can be done, an investigation was made to determine the necessary separation factors. The study indicated

that separations beyond certain limits may not yield enough to substantiate such separation factors. The separations of 99.99% for plutonium, 99.9% for uranium, americium and curium, and 99% for neptunium will reduce the hazard potential to about five percent of that for natural uranium.⁽³⁾ After 99.9% removal of iodine, it will then be the long-lived remaining fission products which control the waste hazard. Higher removal factors for the actinides do not appear to be warranted unless long-lived fission products are also removed, especially Tc-99.

As means of recovering actinides from the spent waste, several schemes are available. Several schemes can be ruled out mainly due to expense and complexity. For example, a centrifuge is too "dirty" because of associated alpha emitters from the actinides.⁽⁴⁾ This would require tight contamination control, and hence much shielding. Other processes require a gaseous form, but there are no gaseous forms of americium or curium.

Present feasibility studies indicate that separations based on solvent extraction, ion exchange, and scavenging precipitation have greatest possibilities. Solvent extraction by itself has not been shown to achieve desired results; however, multi-step solvent extraction processes have a greater probability of success.⁽⁵⁾ If particular waste stream recycles solved, processes based on cation exchange may be a viable method for partitioning the actinides. Another method with potential in waste partitioning may be precipitation.

Figure A.2 illustrates the reprocessing scheme for fission products and actinides generated from Light Water Reactors. Spent fuel from LWRs

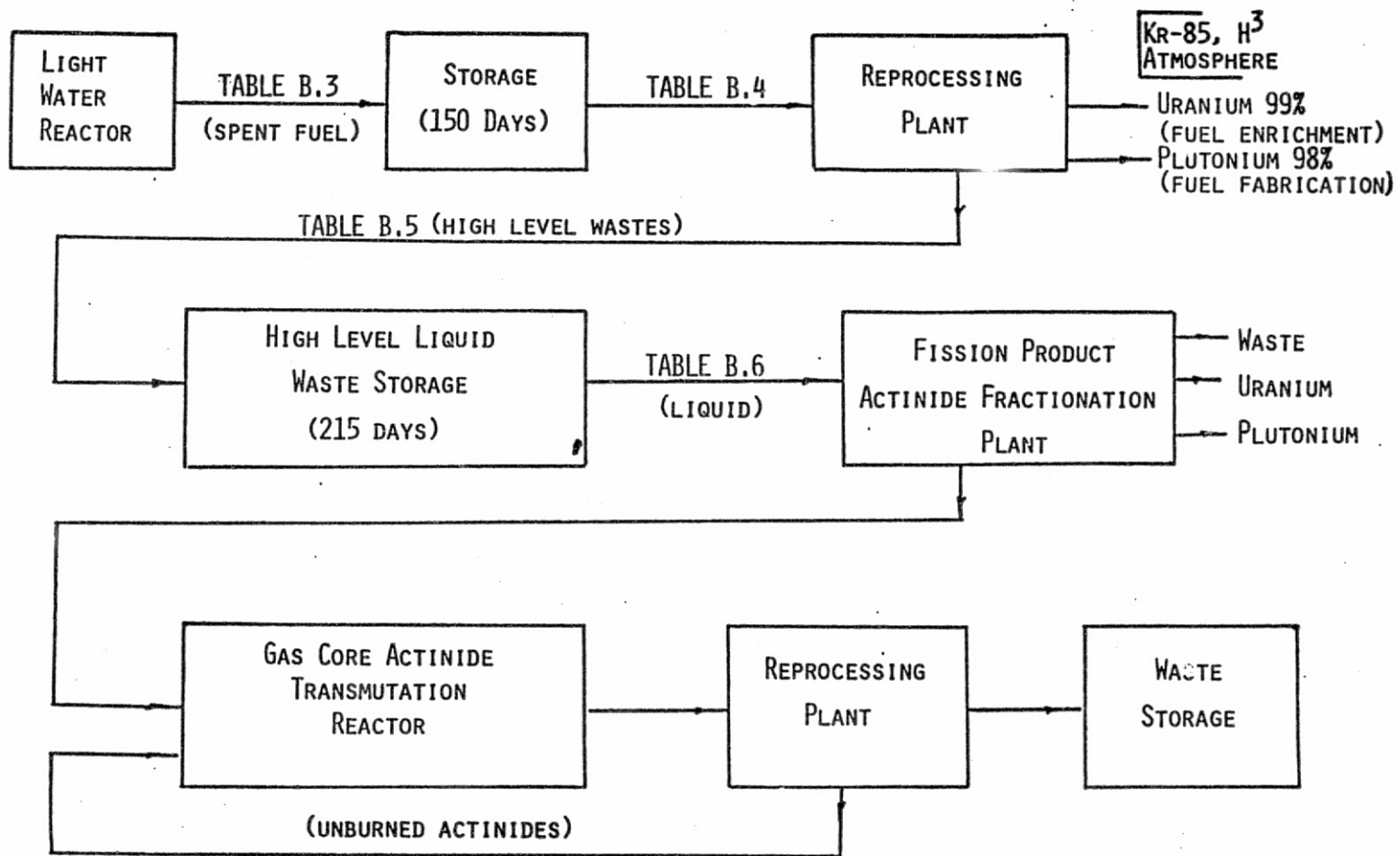


Fig. A.2 Actinide Reprocessing Scheme

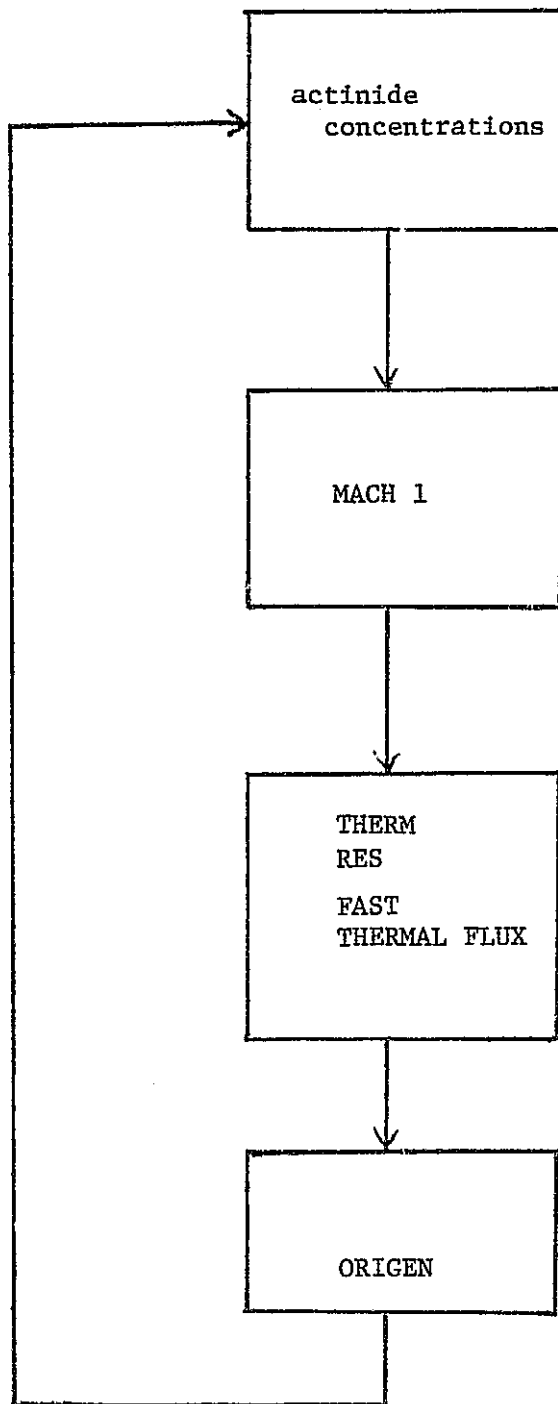


Fig. 4.1. Computational Strategy of UFATR.

containing fission products and actinides listed in Table A.2 is sent to storage for about 150 days. The wastes from storage, which is listed in Table A.3, is then sent to a reprocessing plant. This plant discharges Kr-85 and tritium to the air. Ninety nine percent of the uranium is removed from the waste and sent for enrichment and 98 percent of the plutonium is separated for further fuel fabrication.

The rest of the high-level waste goes to a high-level liquid waste storage for about 215 days. These high-level wastes are listed in Table A.4. After further storage these wastes (listed in Table A.5) go to a fission product/actinide fractionation plant.

Fractionation Schemes

Studies to date indicate that the best methods for removing actinides from wastes will be obtained by improving present state-of-the-art methods.⁽⁶⁾ One of the present schemes is shown in the Fig. A.3. In this scheme, neptunium, uranium, and plutonium, are recovered in the primary PUREX plant. Various exhaustive extractions or further PUREX processes are used to accomplish complete removal of the neptunium, plutonium, and uranium. Through the PUREX plant process, a recovery rate of 95-99% for neptunium and improvements in uranium and plutonium recovery to 99.5% or better are expected.⁽⁷⁾

The interim waste storage is for the purpose of reducing the radiation hazard from the remaining high level wastes during subsequent processing. The radiation hazard will be high unless the fission product yttrium and rare earths, which are associated with americium and curium, are allowed to

TABLE A.2

FISSION PRODUCT AND ACTINIDE CONCENTRATIONS LEAVING A LWR

PWR FUEL CYCLE - DECAY TIMES OF FUEL DURING COOLING PERIOD
 POWER= 30.00MW, BURNUP= 33000.MWD, FLUX= 2.92E+13N/CM**2-SEC
 NUCLIDE INGESTION HAZARD, M**3 OF WATER AT RCG
 BASIS = PER METRIC TONNE OF U LOADED IN REAC

Actinides		Fission Products	
	DISCHARGE		DISCHARGE
PB2112	7.50E+01	H 3	2.36E+05
BI2112	3.75E+01	KR 85	1.13E+04
RA2223	1.27E+02	RB 86	1.47E+07
RA2223	7.50E+02	SR 89	3.39E+11
TH2233	2.13E+02	SR 90	5.59E+11
TH2333	8.88E+00	Y 90	4.03E+09
TH2333	5.93E+03	Y 91	3.13E+10
TH2334	1.57E+04	ZR 93	2.66E+03
PA2231	2.71E+01	NB 93M	3.08E+02
PA2233	5.24E+03	ZR 95	2.29E+10
PA2234M	1.60E+01	NB 95M	2.80E+04
PA2334	6.55E+01	NB 99	1.33E+10
U2332	2.02E+02	MO 99	3.81E+10
U2333	1.52E+00	TC 99	7.14E+04
U2334	2.55E+04	RU103	1.52E+10
U2355	5.70E+02	RH103M	1.22E+08
U2356	9.61E+03	RU106	5.45E+10
U2376	8.53E+09	RH106	7.40E+05
U2378	7.65E+03	PD107	1.10E+02
NP2237	1.11E+05	AG110M	1.23E+06
NP2239	1.83E+11	AG111	1.59E+05
PU2236	1.17E+04	AG111	9.90E+08
PU2239	5.43E+08	CD113M	7.05E+04
PU2239	6.60E+07	IN114M	1.75E+07
PU2240	9.30E+08	CD115M	1.34E+07
PU2241	2.76E+08	SN119M	1.64E+01
PU2242	2.15E+07	SN123	8.33E+03
AM2241	2.22E+06	SB124	2.63E+07
AM2242M	6.84E+08	SN125	6.76E+08
AM2243	4.54E+06	SB125	6.70E+07
AM2243	1.67E+09	TE125M	3.11E+07
CM2243	7.42E+05	TE127M	3.07E+08
CM2244	3.19E+08	TE127	6.60E+08
CM2245	8.54E+04	TE129M	2.46E+08
CM2246	1.71E+04	TE129	4.21E+08
CM2248	1.98E+00	I129	5.18E+05
BK2249	8.96E+00	I131	2.37E+12
CF2250	3.79E+00	XE131M	6.33E+03
CF2252	2.53E+00	TE132	5.92E+10
SUBTOT	1.93E+11	I132	1.53E+11
		XE133	1.61E+06
		CS134	2.74E+10
		CS135	2.36E+03
		CS136	1.01E+09
		CS137	5.39E+09
		BA137M	1.01E+05
		BA141	7.27E+10
		LA140	7.50E+10
		CE141	1.54E+10
		PR143	2.41E+10
		CE144	1.11E+11
		PR144	1.12E+06
		NO147	9.81E+09
		PM147	5.12E+08
		PM148M	6.89E+04
		PM149	9.93E+05
		SM151	3.12E+06
		EU152	1.57E+05
		GO153	4.73E+05
		EU154	6.49E+08
		EU155	6.74E+07
		EU156	5.26E+05
		TS160	3.21E+07
		SUBTOT	4.11E+12
TOTALS	2.16E+11	TOTALS	6.40E+12

TABLE A.3

FISSION PRODUCT AND ACTINIDE CONCENTRATIONS AFTER 150 DAYS STORAGE

PWR FUEL CYCLE - DECAY TIMES OF FUEL DURING COOLING PERIOD
 POWER= 30.00MW, BURNUP= 33000.MWD, FLUX= 2.92E+13N/CM**2-SEC
 NUCLIDE INGESTION HAZARD, M**3 OF WATER AT RCG
 BASIS = PER METRIC TONNE OF U LOADED IN REAC

Actinides		Fission Products	
	150. D		150. D
PB212	1.10E+02	H 3	2.31E+05
BI212	5.49E+00	KR 85	1.10E+04
RA223	1.40E+00	RB 86	9.49E+04
RA224	1.10E+03	SR 89	3.24E+10
TH223	3.10E+02	SR 90	2.36E+11
TH233	1.02E+01	Y 90	3.34E+09
TH231	3.55E+01	Y 91	5.37E+09
TH234	3.57E+04	ZR 93	2.36E+03
PA231	3.74E+01	NB 93M	4.52E+02
PA233	3.46E+03	ZR 95	4.52E+09
PA234M	1.57E+01	NB 95M	4.52E+03
PA234	1.44E+02	NB 95	5.22E+04
U232	1.40E+02	MO 99	2.55E+06
U233	1.10E+00	TC 99	7.17E+04
U234	1.10E+04	RU103	1.10E+09
U235	1.10E+02	RH103M	8.83E+06
U236	1.10E+03	RU106	4.10E+10
U237	1.10E+04	RH106	4.10E+05
U238	7.85E+03	PD107	1.10E+02
NP237	1.10E+05	AG110M	3.14E+07
NP239	1.10E+05	AG111	3.14E+02
PU236	1.10E+04	AG111	9.47E+02
PU238	1.10E+08	CO113M	1.10E+01
PU239	1.10E+07	IN114M	9.62E+03
PU240	1.10E+07	CO115M	1.10E+06
PU241	1.10E+08	SN119M	1.10E+01
PU242	1.10E+05	SN123	3.86E+03
AM241	1.10E+07	SB124	3.86E+06
AM242M	1.10E+06	SN125	1.10E+04
AM242	1.10E+04	SB125	7.99E+07
AM243	1.10E+06	TE125M	3.14E+07
CM242	1.10E+08	TE127M	1.10E+08
CM243	1.10E+05	TE127	3.14E+07
CM244	1.10E+08	TE129M	1.10E+08
CM245	1.10E+04	TE129	1.10E+06
CM246	1.10E+04	I129	1.10E+05
CM248	1.10E+00	I131	1.10E+00
BK249	1.10E+00	XE131M	1.10E+00
CF250	1.10E+00	TE132	1.10E+00
CF252	1.10E+00	I132	1.10E+00
SUBTOT	2.52E+09	XE133	1.10E+00
		CS134	1.10E+10
		CS135	1.10E+09
		CS136	1.10E+09
		CS137	1.10E+09
		BA143M	1.10E+04
		BA143	1.10E+07
		LA143	1.10E+07
		CE141	1.10E+08
		PR143	1.10E+07
		CE144	1.10E+10
		PR144	1.10E+05
		NO147	1.10E+05
		PM147	1.10E+08
		PM148M	1.10E+08
		PM148	1.10E+08
		SM151	1.10E+08
		EU152	1.10E+08
		GO153	1.10E+08
		EU154	1.10E+08
		EU155	1.10E+07
		EU156	1.10E+08
		TB160	1.10E+06
		SUBTOT	4.58E+11
TOTALS	2.52E+09	TOTALS	4.58E+11

TABLE A.3

FISSION PRODUCT AND ACTINIDE CONCENTRATIONS AFTER 150 DAYS STORAGE

PWR FUEL CYCLE - DECAY TIMES OF FUEL DURING COOLING PERIOD
 POWER= 30.00MW, BURNUP= 33000.MWD, FLUX= 2.92E+13N/CM**2-SEC
 NUCLEIDE INGESTION HAZARD, M**3 OF WATER AT RCG
 BASIS = PER METRIC TONNE OF U LOADED IN REAC

Actinides			Fission Products		
	150.	D		150.	D
PB2112	1.16E	+02	H	3	
BI2112	5.49E	+00	KR	85	-
RA2223	1.70E	+00	RB	86	
RA2224	1.1E	+03	SR	89	
TH2223	3.1E	+02	SR	90	
TH233u	1.1E	+01	Y	90	
TH2331	3.5E	+01	Y	91	
TH2334	1.5E	+04	ZR	93	
PA2331	3.7E	+01	NB	93M	
PA2333	3.4E	+03	ZR	95M	
PA2334M	1.5E	+01	NB	95M	
PA2334	1.5E	+01	NB	95M	
U2332	3.5E	+02	MO	99	
U2333	1.1E	+00	TC	99	
U2334	3.5E	+04	RU103		
U2335	3.5E	+02	RH103M		
U2336	9.8E	+03	RU106		
U2337	3.5E	+04	RH106		
U2338	7.8E	+03	PD107		
NP2337	1.1E	+05	AG111M		
NP2339	1.1E	+05	AG111		
PU2335	1.2E	+04	AG111		
PU2338	3.5E	+08	CD113M		
PU2339	3.5E	+07	IN114M		
PU240	3.5E	+07	CD115M		
PU241	3.5E	+08	SN119M		
PU242	3.5E	+05	SN123		
AM241	3.5E	+07	SB124		
AM242M	3.5E	+06	SN125		
AM242	3.5E	+04	SB125		
AM243	3.5E	+06	TE125M		
CM242	3.5E	+08	TE127M		
CM243	3.5E	+05	TE127		
CM244	3.5E	+08	TE129M		
CM245	3.5E	+04	TE129		
CM246	1.7E	+04	I129		
CM248	1.1E	+00	I131		
BK249	3.5E	+00	XE131M		
CF250	3.5E	+00	TE132		
CF252	3.5E	+00	I132		
SUBTOT	2.5E	+09	XE133		
			CS134		
TOTALS	2.52E	+09	CS135		
			CS136		
			CS137M		
			BA143		
			LA143		
			CE141		
			PR143		
			CE144		
			PR144		
			NO147		
			PM147		
			PM148M		
			PM148		
			SM151		
			EU152		
			GO153		
			EU154		
			EU155		
			EU156		
			TB160		
			SUBTOT		
			TOTALS	4.58E	+11

TABLE A.4

FISSION PRODUCT AND ACTINIDE CONCENTRATIONS EXITING FROM THE REPROCESSING PLANT

PWR FUEL CYCLE DECAY TIMES OF FUEL AFTER 1ST PROCESSING
 POWER= 30.00MW, BURNUP= 33000.MWD, FLUX= 2.92E+13N/CM**2-SEC
 NUCLIDE INGESTION HAZARD, M**3 OF WATER AT RCG
 BASIS = PER METRIC TONNE OF U LOADED IN REAC

<u>Actinides</u>		<u>Fission Products</u>	
	DISCHARGE		DISCHARGE
PB212	1.10E+02	H 3	2.31E+05
BI212	5.49E+00	KR 85	1.10E+04
RA223	1.79E+00	RB 86	9.49E+04
RA224	1.10E+03	SR 89	3.24E+10
TH228	3.18E+02	SR 90	2.56E+11
TH230	1.02E+01	Y 90	3.24E+09
TH234	1.57E+04	Y 91	9.27E+09
PA233	2.74E+01	ZR 93	2.35E+03
PA233M	4.46E+03	NB 93M	4.55E+02
U233	2.45E+00	ZR 93M	5.62E+09
U233M	5.52E+02	NB 93M	5.88E+03
U235	9.70E+00	NB 93M	3.20E+09
U235M	9.61E+04	TC 99	7.17E+04
U237	2.65E+02	RU106	1.12E+09
U238	7.85E+01	RH103M	8.33E+06
NP237	1.13E+05	RU106	4.10E+00
NP239	1.82E+05	RH106	4.10E+03
PU238	2.12E+02	PO107	1.10E+02
PU238M	1.13E+07	AG110M	8.14E+07
PU239	1.29E+06	AG110M	8.17E+02
PU240	1.91E+05	CD113M	1.33E+01
PU241	1.03E+07	IN114M	9.59E+03
PU242	3.52E+06	CD113M	4.64E+06
AM241M	2.29E+06	SN119M	1.08E+01
AM242	9.15E+04	SN123	3.30E+05
AM243	4.34E+06	SB124	3.59E+06
AM244	3.88E+08	SB125	7.89E+07
CM243	7.36E+05	TE125M	3.20E+07
CM244	3.44E+08	TE127M	4.23E+08
CM245	2.34E+04	TE127M	3.04E+07
CM246	1.71E+04	TE129M	1.33E+08
CM248	1.90E+00	TE129M	2.17E+06
BK249	6.44E+00	I131	6.23E+09
CF249	8.96E+01	I131	7.28E+06
CF251	13.69E+00	CS134	2.68E+10
CF252	2.36E+00	CS135	2.89E+03
SUBTOT	1.30E+09	CS136	3.41E+09
		CS137	5.64E+09
TOTALS	1.30E+09	BA137M	9.39E+04
		BA140	2.16E+07
		LA140	2.48E+07
		CE141	6.27E+08
		PR143	1.36E+07
		CE144	7.71E+10
		PR144	7.71E+09
		NO147	8.39E+05
		PM147	4.90E+08
		PM143M	3.37E+03
		PA143	2.83E+02
		SM151	3.12E+06
		EU152	1.53E+05
		GD153	1.16E+05
		EU154	3.43E+08
		EU155	3.20E+07
		TD160	7.53E+06
		SUBTOT	4.58E+11
		TOTALS	4.58E+11

TABLE A.5

**FISSION PRODUCT AND ACTINIDE CONCENTRATIONS AFTER 215 DAYS
STORAGE IN HIGH LEVEL LIQUID WASTE STORAGE FACILITY**

PWR FUEL CYCLE DECAY TIMES OF FUEL AFTER 1ST PROCESSING
POWER= 30.00MW, BURNUP= 33000.MWD, FLUX= 2.92E+13N/CM**2-SEC
NUCLIDE INGESTION HAZARD, M**3 OF WATER AT RCG
BASIS = PER METRIC TONNE OF U LOADED IN REAC

<u>Actinides</u>			<u>Fission Products</u>		
	CHARGE	215. D			215. D
PB212	0.	9.11E+01	H	3	2.23E+05
BI212	0.	4.55E+01	KR	85	1.06E+04
RA223	0.	2.33E+00	RB	86	3.28E+01
RA224	0.	9.11E+02	SR	89	1.84E+09
TH228	0.	2.59E+02	SR	90	2.52E+11
TH230	0.	1.02E+01	Y	90	3.79E+09
TH234	0.	1.89E+02	Y	91	4.26E+08
PA231	0.	2.74E+01	ZR	93	2.36E+03
PA233	0.	3.40E+03	NB	93M	5.78E+02
U232	0.	3.56E+00	ZR	93	4.67E+08
U234	5.45E+04	2.56E+02	NB	95M	5.94E+02
U235	2.36E+03	9.61E+00	NB	95	5.96E+08
U236	0.	9.61E+01	TC	93	7.17E+04
U237	0.	4.81E+02	RU	103	2.56E+07
U238	8.05E+03	7.85E+01	RH	103M	2.05E+05
NP237	0.	1.13E+05	RU	106	2.73E+10
NP239	0.	1.82E+05	RH	106	2.73E+05
PU236	0.	1.84E+02	PD	107	1.10E+02
PU238	0.	2.19E+07	AG	110M	4.51E+07
PU233	0.	1.29E+06	AG	110	1.76E+02
PU240	0.	1.94E+06	GD	113M	9.99E+00
PU241	0.	1.00E+07	IN	114M	4.92E+02
PU242	0.	5.52E+03	CU	115M	5.11E+04
AM241	0.	3.90E+07	SN	113M	5.96E+00
AM242M	0.	2.28E+06	SN	123	1.17E+03
AM242	0.	9.12E+04	SB	124	2.99E+05
AM243	0.	4.54E+06	SB	125	6.83E+07
CM242	0.	3.56E+08	TE	125M	2.83E+07
CM243	0.	7.26E+05	TE	127M	3.13E+07
CM244	0.	3.36E+08	TE	127	7.74E+06
CM245	0.	8.54E+04	TE	129M	1.69E+06
CM245	0.	1.71E+04	TE	129	2.71E+04
CM248	0.	1.98E+00	I	123	6.24E+05
CK243	0.	4.01E+00	I	131	6.64E+02
CF243	0.	1.49E+00	CS	134	1.95E+10
CF250	0.	3.58E+00	CS	135	2.86E+03
CF252	0.	1.94E+00	CS	136	5.58E+00
SUBTOT	6.50E+04	7.74E+08	CS	137	5.27E+09
			BA	137M	3.85E+04
TOTALS	6.50E+04	7.74E+08	BA	140	1.89E+02
			LA	140	2.18E+02
			CE	141	6.31E+06
			PR	143	2.56E+02
			CE	144	4.56E+10
			PR	144	4.56E+05
			ND	147	1.24E+00
			PM	147	4.19E+08
			PM	148M	9.40E+01
			PM	148	7.55E+00
			SM	151	3.11E+06
			EU	152	1.48E+05
			GD	153	6.25E+04
			EU	154	3.35E+08
			EU	155	2.55E+07
			TB	160	9.60E+05
			SUBTOT		3.56E+11
			TOTALS		3.56E+11

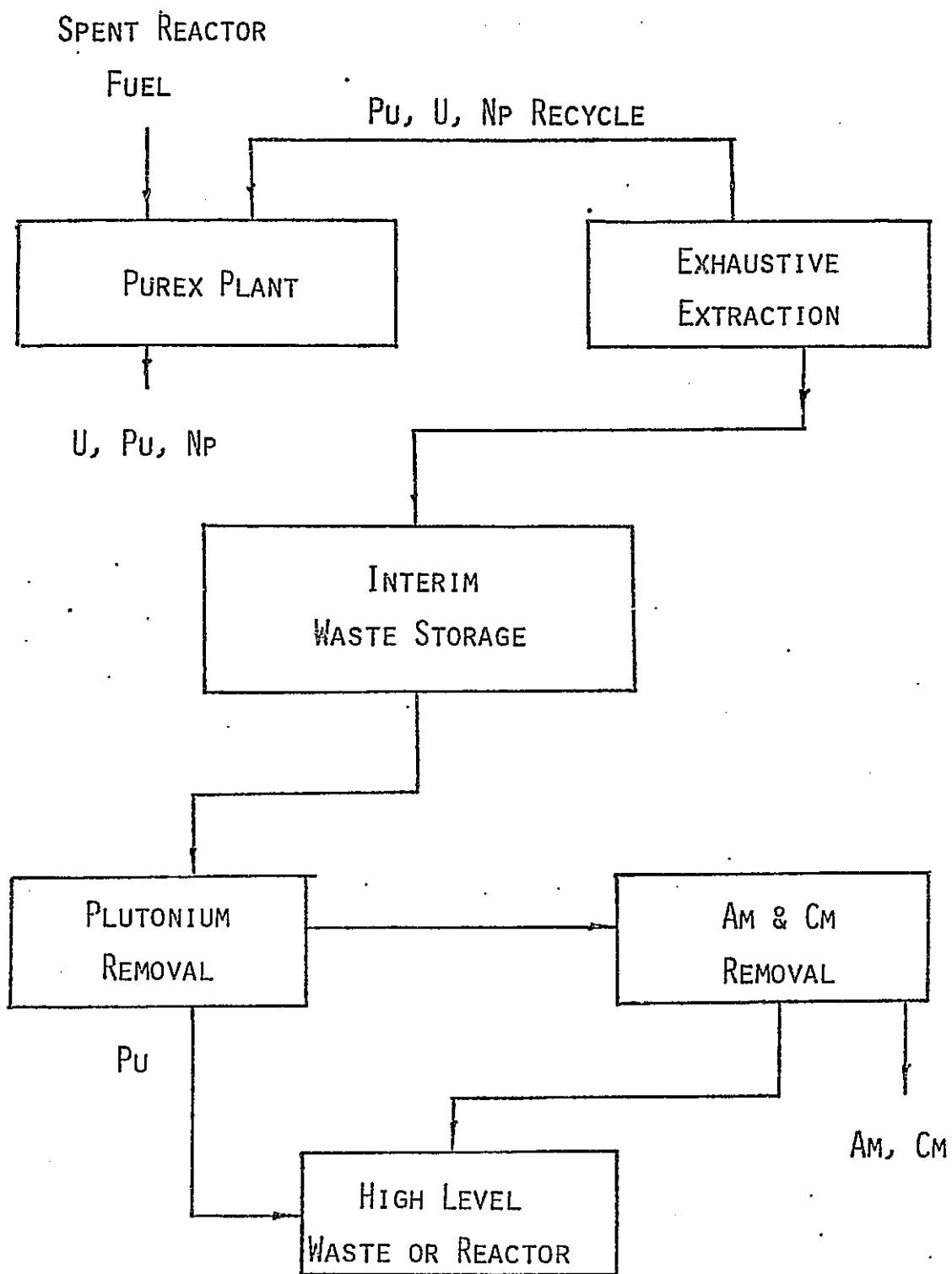


Fig. A.3 Present Processing Sequence for the Removal of Actinides

decay to less hazardous levels.⁽⁷⁾ By considering the most important decay times, storage times of ten years would significantly reduce the hazards. Current NRC regulations require that wastes be solidified within five years. However, because of difficulties in working with a solid waste, it will be assumed that the americium and curium are removed from the liquid wastes after a five year period.

One disadvantage of interim waste storage is that the amount of plutonium in the waste grows by curium decay. Therefore, plutonium removal from the stored waste is necessary after several years of interim storage. The process showing most potential for recovering the plutonium is an all ion-exchange process.⁽⁸⁾

After removal of plutonium, the americium and curium are isolated from the rest of the waste. The problems associated with americium and curium removal are centered around finding a suitable chemical separation process for commercial high level wastes. Recovery of americium and curium has been done at the Oak Ridge National Laboratory and Savannah River Laboratory on a multigram basis using a Tramex process.⁽⁷⁾ This process has problems with corrosive solutions that require processing equipment constructed of special and expensive materials. Because of these reasons, the process is not recommended. However, there is some possibility that the Tramex processing equipment can be constructed so as to allow safe working of both corrosive solution in the process and toxic radionuclides at little additional cost.

Other processes that have been developed and claim to give high americium and curium separation are Cation Exchange Chromatography (CEC)

and Trivalent Actinide-Lanthanide Separation by Phosphorous Reagent Extraction from Aqueous Complexes (TALSPEAK).⁽⁷⁾ Cation Exchange Chromatography was developed at the Savannah River Laboratory and successfully used to separate about twenty-five percent of the necessary amounts of americium, curium, and rare earths in one metric ton of Light Water Reactor fuel.⁽⁷⁾ A schematic flowsheet of CEC is shown in Fig. A.4. The TALSPEAK process, shown in Fig. A.5, has been developed only to the point of tracer-level laboratory studies at Karlsruhe for americium and curium removal.⁽⁷⁾

As means of separating Am and Cm from other wastes, the Tramex, CEC, and TALSPEAK processes require considerable developmental work and data gathering to determine their applicability to the commercial (high volume) extraction of actinides from high-level wastes.

Proposed Schemes

Present proposals for actinide partitioning are based on a sequence of separation processes using solvent extraction, ion exchange, and precipitation. These techniques have not yet been developed.⁽⁶⁾ A multistep solvent extraction process combined with other processes, such as cation exchange, may work well in the removal of uranium, neptunium, and plutonium, as well as separations of americium and curium from other wastes.

Tributylphosphate (TBP) may be used as the solvent in the solvent extraction method.^(6,9) As demonstrated in the PUREX process, TBP achieved highly efficient recovery of uranium, plutonium, and neptunium.⁽³⁾

As a means of separating americium and curium from the rest of fission products and wastes, two steps of cation exchange is quite promising. The potential here appears to be 99.9 percent or better.⁽⁶⁾ In the first step

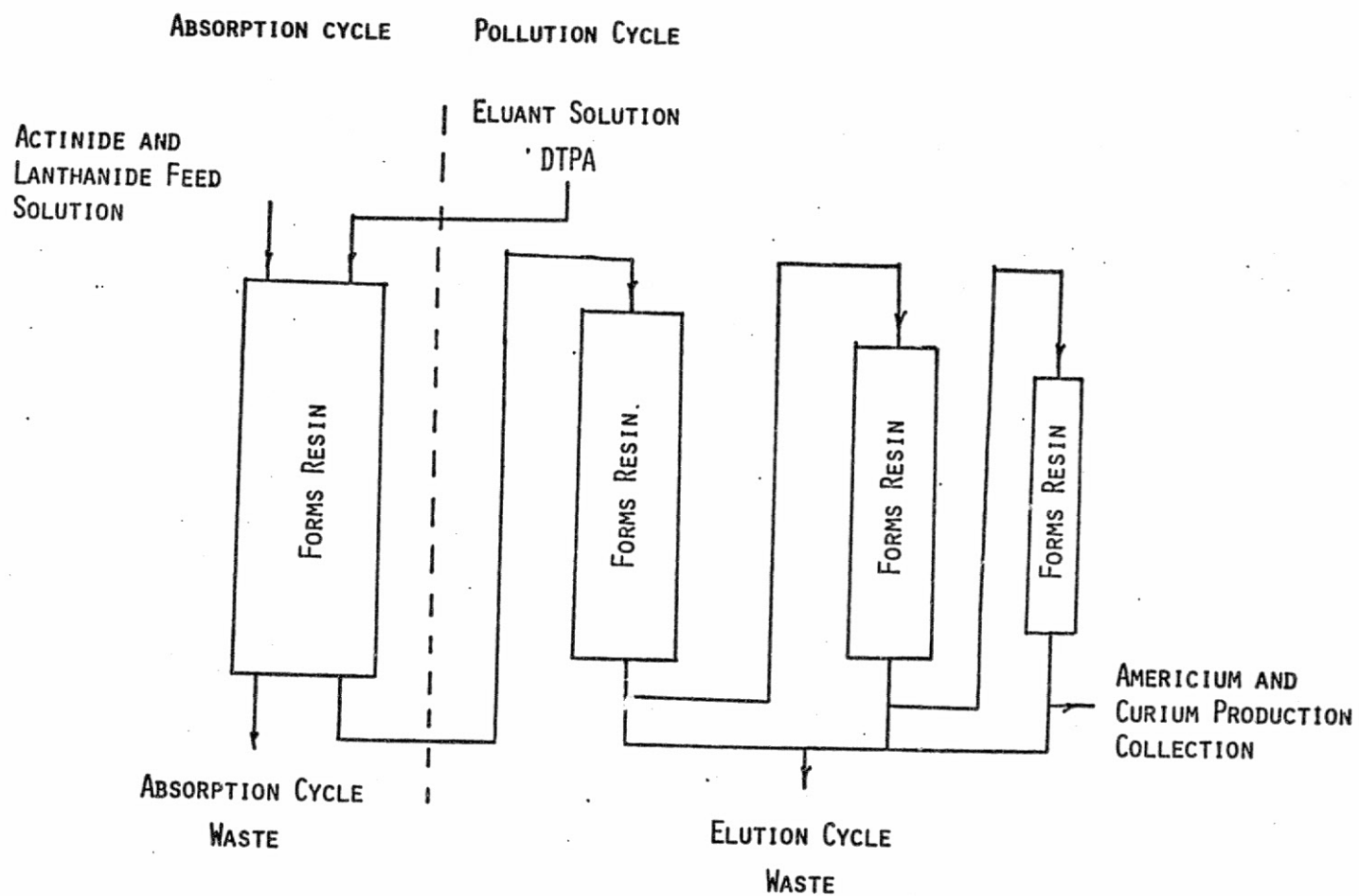


Fig. A 4 Schematic Flowsheet of Cation Exchange Chromatographic Process for Recovery of Americium and Curium⁽¹⁵⁾

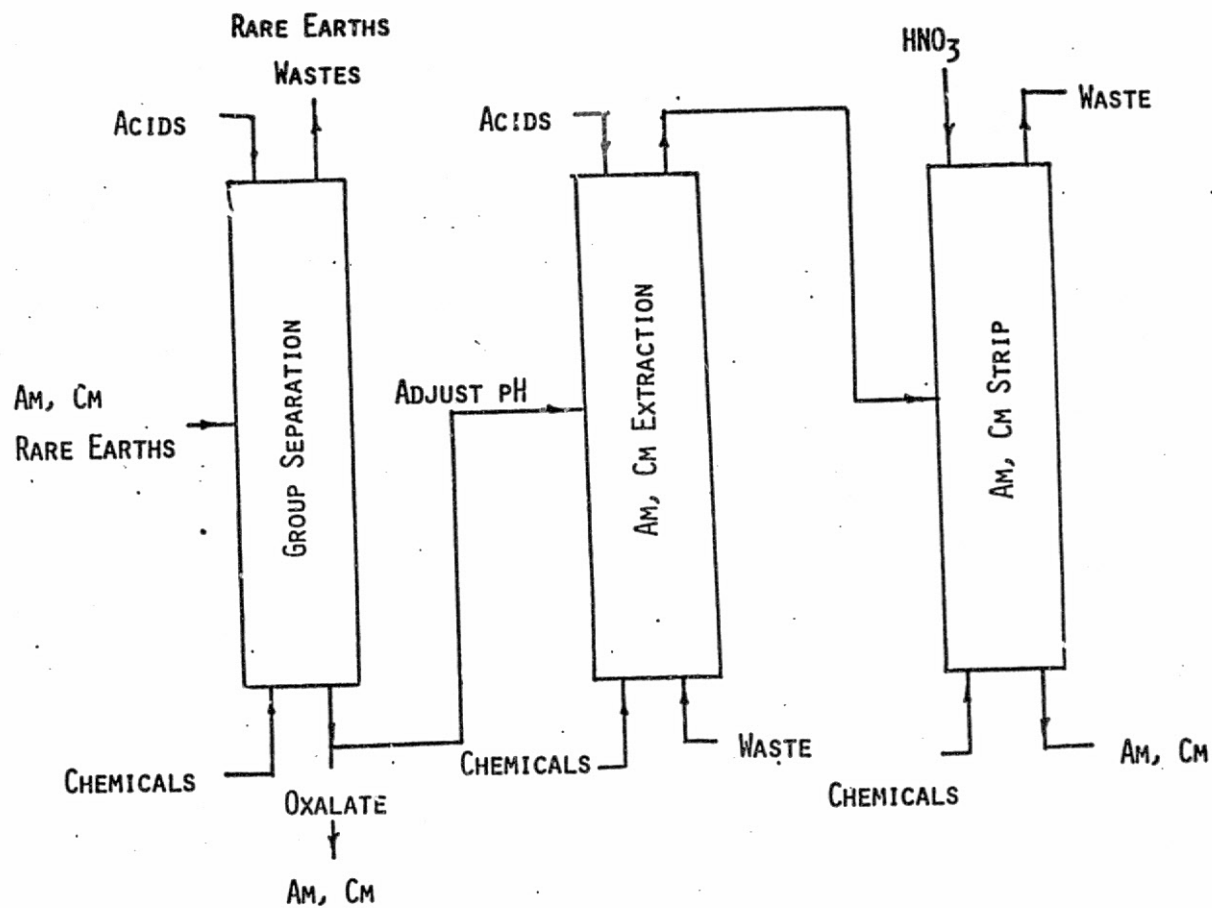


Fig. A.5 Conceptual Flow Sheet for Recovery of Americium and Curium by a TALSPEAK

the lanthanides and actinides are absorbed on a cation exchange resin column and eluted with nitric acid. In the following step the lanthanides and actinides are separated by cation exchange chromatography. Problems to be solved with this process are in converting the spent ion exchange resin to acceptable levels for waste generated in the chromatographic separation.

Precipitation methods combined with ion exchange and/or solvent extraction may be another possible method for partitioning actinides. Even though solid waste handling is unavoidable, ways are now under study for obtaining crude concentrations of plutonium, americium, curium, and fission products. These actinides would then be separated from the lanthanides in further ion exchange or solvent extraction steps. Oak Ridge National Laboratory is studying the use of oxalate⁽⁸⁾ precipitation together with ion exchange to isolate the lanthanides and actinides.^(6,11) A removal factor of 0.95 is achieved by precipitation while the remaining is removed in the cation exchange column.⁽⁷⁾ Tracer-level studies indicate removal of 0.999 for americium and curium.⁽⁷⁾ Almost complete removal has been demonstrated for americium and curium by use of multiple oxalate precipitation stages.⁽⁶⁾ Further work in this area is still needed to determine the effect of the handling problems.

Technical feasibility, resultant benefits, and costs of partitioning actinides from high-level wastes are yet to be established. It must be decided if the net benefits will justify the use of partitioning. It must also be kept in mind that the separation schemes do not solve the long-term actinide problem. In order to justify this, the actinides

must somehow be transmuted to shorter-lived radionuclides or disposed of from our environment. These and many more problems still need research and investigation before a feasible actinide-separation-transmutation process can be substantiated.

From research done to date, it is concluded that much research and development is still needed in the area of actinide partitioning. Work being performed at the Oak Ridge National Laboratory may show encouraging results in the near future. Present state-of-the-art methods will not yield the results needed to establish a practical, economically feasible operating partitioning plant. It is believed that research in the area of combined methods of solvent extraction and ion exchange will yield the necessary separations factors.

References for Appendix A

1. Lowry, L. L., "Gas Core Reactor Power Plants Designed for Low Proliferation Potentials," LA-6900-MS (September, 1977).
2. Clement, J. D. and Rust, J. H., "Analysis of the Gas Core Actinide Transmutation Reactor (GCATR)," Annual Report, Georgia Institute of Technology, NASA Grant NSG-1288 (February, 1977).
3. Claiborne, H. C., "Effect of Actinide Removal on the Long Term Hazard of High-Level Waste," ORNL-TM-4724 (January, 1975).
4. Schneider, A., Georgia Institute of Technology, Personal consultation (April, 1976).
5. Bocola, W., Frittelli, L., Gera, F., Grossi, G., Moccia, A., and Tondinelli, L., "Considerations on Nuclear Transmutation for the Elimination of Actinides," IAEA-SM-207/86.
6. Blomeke, J. O., "Technical Alternatives Documents," ORNL, Prepublication Paper (1976).
7. Bond, W. D., and Leuze, R. E., "Feasibility Studies of the Partition of Commercial High-Level Wastes Generated in Spent Nuclear Fuel Processing: Annual Progress Report for FY-1974-1974," ORNL-5012 (January, 1975).
8. Bond, W. D., Claiborne, H. C., and Leuze, R. E., "Methods for Removal of Actinides from High-Level Wastes," Nuclear Technology, 24, 367 (1974).
9. LaRiviere, J. R., et al., "The Hanford Isotopes Production Plant Engineering Study," HW-77770, Hanford Atomic Products Operation (July, 1963).
10. Rupp, A. F., "A Radioisotope-Oriented View of Nuclear Waste Management," ORNL-4776 (May, 1972).
11. Ferguson, D. W., et al., "Chemical Technology Division Annual Progress Report for Period Ending March 31, 1975," ORNL-5050, 6-11, 30-31 (October, 1975).